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DATA ON THE PHOSPHORUS CONTENT AND ORGANIC REMAINS OF MANGANESE OXIDE ORES FROM URKUT

(Preliminary report)

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INTRODUCTION

The phosphorus content of the manganese oxide ores, beyond certain limites, is undesirable from certain practical point of view, e. g. from that of the producing of ferromanganese. From that point of view it is interesting to examine the phosphorus content and its possible regional changing in the manganese oxide ores of Urkut. Further, also the study of the organic matter may results data completing our knowledge on the age and circumstances of formation of these manganese oxide ores.

Here below these two problems will be briefly discussed, the complete explanation of the problems mentioned above will be given only after bringing to an end the examinations under way.

THE PROBLEM OF THE PHOSPHORUS CONTENT OF THE ORES

In connection with the phosphorus content the following questions may arise:

a) whether or not a connection can be established between the change of the phosphorus content and the ore content of the samples, whether the phosphorus content is accompanied with the ore or with the gangue?

b) whether or not a regularity can be established in the distribution of the phosphorus content?

c) what mineral is the phosphorus-bearing one?

The distribution and changing of the phosphorus content

The phosphorus content of the different samples taken from various places of the mine is shown in Table 1. Therein are also comprised the percentual amounts of the following components: SiO_2, Fe_2O_3, Al_2O_3, $MnO_2 + MnO$, CaO.

Table 1.

No. of the samples	Site of the sampling	SiO_2	Al_2O_3	Fe_2O_3	MnO_2 + MnO	CaO	P_2O_5
		weight per cent					
1	Shaft II,	22,22	7,28	29,85	25,84	1,92	0,39
1/a	„	13,44	3,30	9,72	56,37	2,28	0,26
2	„	24,33	2,12	15,45	41,46	2,33	0,57
3	„	15,20	2,11	9,06	58,84	2,61	0,07
3/a	„	13,67	3,24	20,13	45,84	2,61	0,91
4	„	57,82	7,48	11,56	9,39	1,54	0,55
5	„	11,13	4,13	20,90	49,70	1,91	0,45
6	„	10,22	6,69	16,43	50,28	2,98	1,08
7	„	19,77	5,20	6,86	45,56	3,63	0,10
8	„	13,24	1,94	8,25	58,38	1,90	0,62
9	„	19,10	1,07	14,94	40,16	1,64	0,45
10	„	3,65	1,38	5,96	75,51	2,02	0,45
11	„	10,09	4,47	26,11	41,15	2,73	0,94
12	Shaft III,	12,00	5,03	16,43	49,80	1,87	0,10
13	chute 4	6,13	4,69	17,43	59,54	1,19	0,18
14	„	43,87	24,51	14,82	3,56	1,30	0,03
15	„	6,81	4,67	19,42	47,43	1,32	0,08
16	„	7,49	14,54	25,56	35,55	1,33	0,04
17	„	65,19	9,60	9,50	5,93	1,33	0,02
18	Shaft III,	27,00	1,63	3,01	35,17	12,39	7,97
19	chute 2	7,99	5,19	9,60	57,82	2,69	0,35
20	Mt. Csárda	1,66	3,09	37,43	45,34	1,27	0,31
21	„	34,50	22,61	24,42	0,53	2,21	0,63
22	„	30,32	25,27	29,17	0,55	1,78	0,61
23	„	6,80	2,30	65,15	13,42	1,05	0,34
24	„	7,51	3,59	56,45	20,05	1,22	0,44
25	„	43,24	17,92	9,25	3,98	2,69	2,39
26	„	55,19	22,29	9,65	0,85	0,97	0,13
27	„	21,54	12,13	18,44	34,08	2,14	0,42
28	„	50,43	15,19	14,58	0,87	3,14	0,66
29	„	52,06	4,08	24,68	14,79	0,99	0,21
30	Hoisting slope	3,99	2,50	10,93	69,23	2,43	0,28

As the manganese oxide content of the samples can be dissolved with dilute hydrochloric acid + hydrogen hyperoxyde treatment, the solubile part of the ores have been dissolved in such a way and the phosphorus content of these solutions have been determined too. The results are given in Table 2.

Table 2.

No. of the samples	P_2O_5 % in the samples	P_2O_5 % in the dissolved part of the samples
2	0,57	0,57
3/a	0,91	0,91
11	0,94	0,84
12	0,10	0,10
13	0,18	0,15
14	0,029	0,02
15	0,08	0,02
16	0,046	0,03
17	0,02	0,005
18	7,97	7,26
19	0,35	0,22
21	0,63	0,52
22	0,61	0,17
23	0,34	0,18

From the results above-mentioned at first sight it could be concluded that significant part of the phosphorus content is associated with manganese oxide content of the samples. The phosphorus content, however, does not change proportionately with the manganese oxide content — as it can be stated from Table 1 —, further, there are clayey samples with no or minimal manganese oxide content, but with considerable phosphorus content and on the other hand, high grade ore samples with relatively minimal phosphorus content can also be found. Thus, the data of Table 2 show at the most only that dissolving the manganese oxide components (and the hydrous iron oxides too) of the samples by treating with hydrochloric acid + hydrogen hyperoxyde, the phosphorus-bearing mineral also dissolves in a considerable amount.

Considering Table 1 it can not be stated that the change of the amount of some components would be proportionate with the change of the phosphorus content.

Hence, the first question in connection with the phosphorus content may be answered: on the basis of the data of analyses can not be concluded that the phosphorus content would be associated with the ore components of the samples.

The data of the Table 1, however, show some regional relations and connections regarding the change of the phosphorus content, though it can not be yet stated that the phosphorus content is higher or lower rather in the samples containing more ore or in the clayey specimens due to some regularity.

Namely, comparing the average P_2O_5 content of the samples from different sampling places, the following picture is obtained shown in Table 3.

Table 3.

Sampling place	Number of samples	P_2O_5 % min.	max.	av.
Shaft II,	13	0,07	1,08	0,52
Shaft III; chute 4	6	0,02	0,18	0,07
Mt. Csárda	9*	0,13	0,66	0,41

* Omitting the solely extremely high value of 2,39 per cent.

On the basis of the above data it seems that the average P_2O_5 content is the lowest in the samples from the area of shaft III, chute 4, whereas those of the samples from shaft II and Mt. Csárda (open-cut mining) are similar with slight deviation, but six — seventhfold of the lowest average value.

The possibility of regional distinguishing of certain sampling places with lowest P_2O_5 content requires further investigations to establish whether or not the area, in the neighborhood of chute 4, shaft III, with extremely low P_2O_5 content but relatively high manganese content, could be delimited horizontally and vertically and what quantity of ore of lowest P_2O_5 content represents etc.

The phosphate mineral of the ores

The assumption that the phosphorus is present as calciumphosphate, is at hand without any special investigations, however, to support this supposition and to identify the mineral itself, would be more circumstantial without a lucky specimen investigated, since there is no unequivocal relation between the change of the P_2O_5 and the CaO content as the latter may be present not only as the component of the apatite, hence, its total amount and its change, respectively, is not necessarily proportionate with the change of the P_2O_5 content.

The P_2O_5 content determined in sample No. 18, is relatively very high: 7,97 per cent. Similarly the CaO content is high: 12,39 per cent, related to the CaO content of the other samples. The F content of sample No. 18 was also determined: 0,70 per cent. On the basis of the ideal composition of fluorapatite, 0,71 per cent F would correspond to 7,97 per cent P_2O_5 determined in the sample. Thus, the phosphorus is presumably present in the sample as fluorapatite. These values would mean that the apatite content of sample No. 18 is about 20 per cent. It seems to be worthwhile to establish by further sampling in the vicinity of the sampling place No. 18 whether this substance with relatively high apatite content forms a thicker or thinner layer or represents a larger or smaller mass etc.

A further question may be the problem of the distribution and the origin of the fluorapatite in the manganese oxides ores of Urkut. The observations and data obtained make the assumption probable that one

part, and perhaps the more considerable part of the phosphorus, is of organic origin and it is transformed secondary to calciumphosphate, apatite. The other part at the same time is possibly present as the apatite of former rocks. The secondary apatite formed by the action of organisms or partly also by inorganic processes is very finely distributed, perhaps microcrystalline, microscopically unobservable. The very fine distribution of the apatite, considered secondary, is possible supported also by that observation that on concentrating the ores, sometimes the phosphorus content of the contentrate increases related to those of the rough ores.

In the section made from sample No. 18 very small needlets could be detected which probably can be identified with the primary apatite needles. These needles, however, occur so scarcely in the section that they can not result the considerable apatite content of this sample, therefore, it is presumed the more considerable part of the apatite to be present in very fine distribution.

THE PROBLEM OF THE ORGANIC MATTER

The chemical examinations of the manganese oxide ores from Urkut showed that the presence of organic matter must be taken into account in several samples.

This statement could be proved both by microscopical and pollen-analytical investigations too. In thin sections of the sample No. 18 algae and hypha threads, further some indeterminable organic matter could be noted. In the preparates made by customary pollen-analytical method some pollen species could be well identified as those of Cingulatisporites, Eucomiidites troedssonii, Striatopollis, Quercus, Castanea, Amaranthaceae as well as a certain sporomorpha with well preserved contour lines, further also parenchyma and epidermis relicts were recognizable besides organic matter of unknown origin. The pollen-analytical investigation was carried out by Mr. *P. Simoncsics.*

It is to be remarked that if in the presence of organic matter the CO_2 content of the samples is determined by igniting the samples in air-current at about 1000° C and absorbing the evolving CO_2 by soda-lime absorptions tube, the result will be the sum of the carbonate CO_2 and the CO_2 corresponding to the C content of the organic remains and derived from the latter by oxydation during the heating in air-current. Thus, using the above-metioned method for CO_2 determination, without considering whether or not besides carbonates also organic remains are present, the amount of the carbonates calculated on the basis of the CO_2 content determined, may be sometimes too highly estimated not corresponding to the actual state. Further not alone the

manganese oxides of higher valency but also the presence of organic matter make questionable the exactness of the FeO determination. Finally, the determination of the pollen species found in the ores can furnish data to complete our present knowledge on the age of the deposition of the single sedimentary layers.

Thus, the comprehensive chemical and pollen-analytical study — with special respect to the organic remains — of the manganese oxide ores from Urkut seems to be required. The investigations are in progress.

LUDWIGITE FROM OCNA DE FER (VASKŐ, BANAT, RUMANIA)

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One of the most interesting minerals of the classic contact region of the Rumanian Banat is ludwigite which was discovered there. Ludwigite forms between Vaskő (Ocna de Fer formally Moravica) and Dognácska (Dognacea) at the foot of the Mountain Dániel in the magnetite between the old mine of Reichenstein, Jupiter and Archangel at a length of 300 m lenselike embeddings, scattered patches and irregular bands. In the mine district of Archangel along the fissures and layers of crystalline limestone, recrystallized to marble of Mezozoic age, the magnetite veins also contain nests, veins and smaller patches of ludwigite.

In larger masses the ludwigite is very finely fibrous. The fibres varying in length form concentrically radiated, or fanlike aggregates starting from the centre. The aggregates are black, silky and have a lustre resembling astrakhan. The length of the single fibres may amount to some cm. The fibres of the aggregates, interwoven with those of the aggregates in the vicinity, form entangled feltlike, very tough masses. The coarsely fibrous masses are more rare where the interwoven dense aggregate of the 5—6 cm long and about 1 mm thick fibres arranged radially or fanlike resemble delusively to the dense black turmaline (schörl) occurrences.

The coarser ludwigite fibres are not monocrystals but aggregates of parallel grouping of very fine fibres varying in length according to the *c* crystallographic axis. The growth of the parallel groupings of fibres often stops and then continues in an oriented manner. The place of the interrupted growth is shown by the separation of the fibres vertically onto the *c* crystallographical axis. Such separations, which are never sharp and do not indicate a cleavage direction, may be often seen an thicker ludwigite fibres, their weathering starts along these directions. Far thinner ludwigite fibres not showing separation directions are still quite intact when the weathering of the thicker fibres

already starts along the separation lines (Photo 1). Both the very fine fibres and the thicker ones (crystal bundles) are very often curved (Photo 2). The fine fibrous masses are extremely tough, the coarser fibres are rigid, their surface of fracture is irregular and of greasy submetallic lustre.

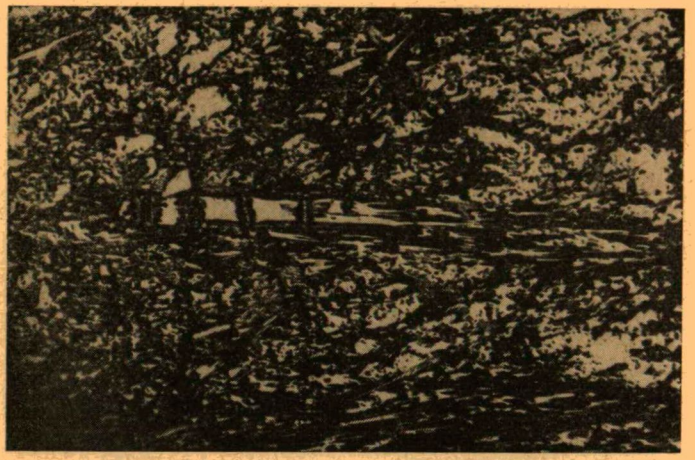

Fig. 1. Coarser ludwigite thread showing separation-lines. The weathering begins along these directions, Plain light, x100.

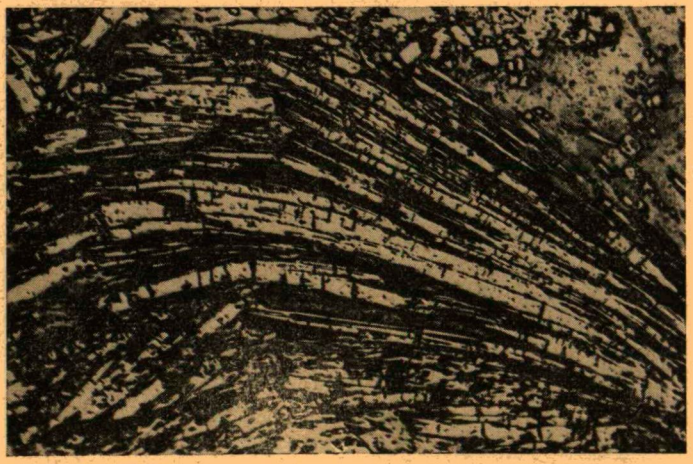

Fig. 2. Curved threads of ludwigite. Plain light, x50.

Generally the ludwigite masses are embedded in magnetite. More rarely one can also find smaller fibrous needle aggregates in marble. The ludwigite fibres reaching into calcite granules which show beautiful polysynthetic twin lamellae, branch into fine needles (Photo 3). Beside the marble, green contact serpentine is also present as an

accessory rock associated with the magnetite-ludwigite ensemble. Very fine ludwigite needles dissolved with dilute HCl from marble show a wonderful pleochroism: bluish green - dark oil green - pinkish light brown, brown. The intensity of the colours depends upon the thickness of the fibres. On the ludwigite needles terminal planes cannot be de-

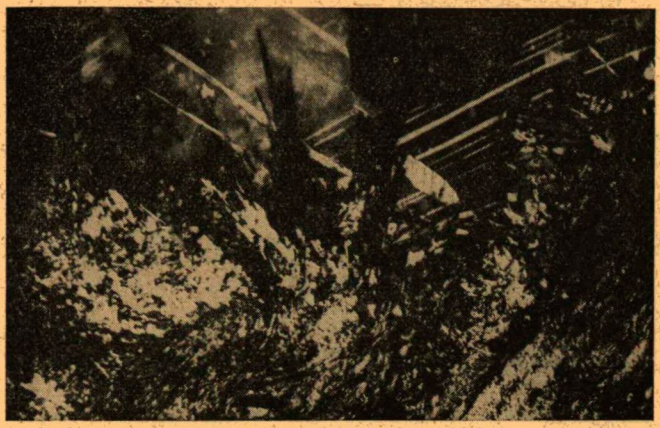

Fig. 3. Ludwigite threads branched into fine needles embedded in calcite. Crossed nicols, x50.

tected, the end of the fibres is irregular i. e. they separate into still finer needles.

The round, small, scattered particles of larger magnetite granules surrounded and replaced by ludwigite can very often be found in the fine-threaded ludwigite masses mostly coeval but partly younger than the magnetite (Photo 4).

Fig. 4. Rounded grains of magnetite replaced by ludwigite. Plain light, x50.

Beside the dominating magnetite and ludwigite, the granules not exceeding 0,1 mm in size or rarely the well developed octahedrons of the younger franklinite can be found overgrown on the edges of the ludwigite fibres or at the separation lines of the fibre-bundles. Its amount is insignificant. Of the sulphide ores pyrite and sphalerite could be observed replacing magnetite and ludwigite (Photo 5).

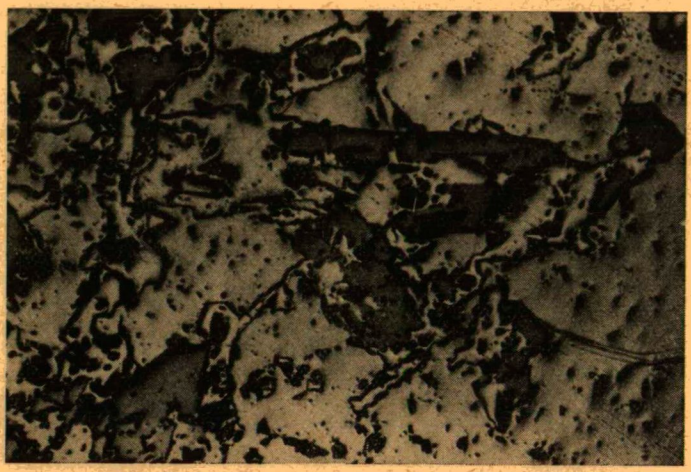

Fig. 5. Ludwigite (gray) replacing sphalerite (white) and pyrite (white with strong relief) as well as magnetite. Plain light, x100.

The dense fine-threaded ludwigite can be well ground and the aggregates perpendicular to the *c* crystallographical axis can excellently be polished, the parts parallel with the longitudinal direction

Fig. 6. Aggregates of ludwigite crystals perpendicular to c axis surrounded by fine threaded ludwigite parallel with the c axis. Crossed nicols, x50.

of the fibres have a duller silky lustre, in this direction the breaking off of the fibres is frequent. Parallel to the longitudinal direction of the fibres, the section is a partly finer and partly coarser fibrous, whereas perpendicular to the *c* axis it shows a »pavement«-structure (Photo 6).

Under the microscope the magnetite is far darker than the ludwigite, beside it magnetite shows a slightly brownish white colour. Franklinite is white still lighter than magnetite. Beside sphalerite, the ludwigite is dark, the magnetite is lighter with a light pinkish shade.

The reflection pleochroism of ludwigite is significant. Parallel to the *c* crystallographical axis it is slightly pinkish lighter or darker grey, perpendicularly to the *c* crystallographic axis it is darker grey — slightly greenish dark grey. In oil the colours are a little more intensive.

Under crossed nicols the extinction is parallel, the interference colors are vivid and change strongly with the orientation. Parallel to the crystallographic axis *c* the color is always pinkish yellowish brown, brown, on the sections perpendicular or nearly perpendicular to the crystallographic axis *c* the colors are far darker: dark bronze red — violet red — bluish red. In oil the intensity of the colors is a littler duller.

Internal reflexes can never be observed. The dark red internal reflexes, mentioned in the Lehrbuch der Erzmikroskopie (Berlin 1931. Bd. II. S. 567.) by *Schneiderhöhn-Ramdohr*, originate from the extremely small franklinite crystals associated with ludwigite.

Etching with ccHNO_3, aqua regia, ccHCl, ccH_2SO_4 is negative. On dropping ccHCl onto the section it shows a mildly green color.

Its structure is always fibrous, the fine fibres compose a dense feltlike network. On the section prepared from coarser fibres, several fibres show a rhombic cross section perpendicular to the crystallographical axis *c*, the obtuse angle of the rhombus measured under the microscope is 113°. Hence the dominating form of the crystal is prism (110).

The result of the analysis of the sample:

MgO	28,55 per cent
FeO	16,39
Fe_2O_3	36,06
MnO	trace
B_2O_3	(17,50)
H_2O	1,50
	100,00 per cent

(Analyst Mrs. Klivényi)

The analysis was prepared from coarsely fibrous substance, the B_2O_3 content is calculated from the difference.

Franklinite and kotoite of pneumatolytic origin are associated in traces with ludwigite. The occurrence of franklinite is not surprising considering that from the ludwigite of this region the Mn could be detected at least in traces, whereas the Zn appears as sphalerite in the

hydrothermal phase. The franklinite is overgrown on the edges of the ludwigite fibres and the aggregate of their extremely small granules quasi rims the ludwigite. Its tiny octahedrons with an adamantine lustre are overgrown on thin ludwigite needles which reach into small hollows. In section, under crossed nicols, it shows a white color and a stronger lustre than magnetite, but particulary in oil it has dark red internal reflexes. It is isotrop. Qualitative determinations showed that it contains both zinc and manganese.

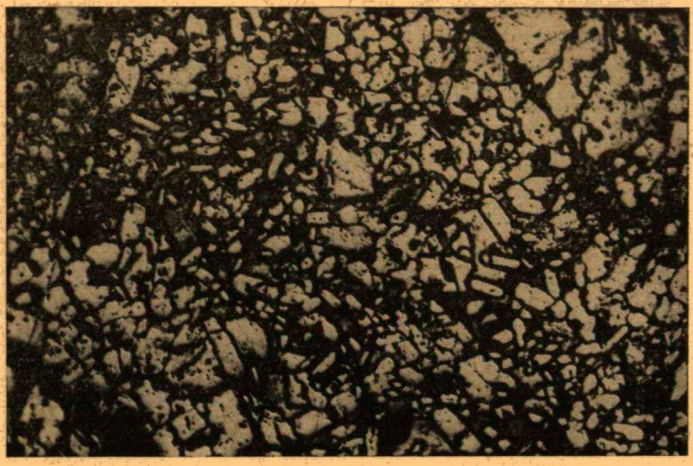

Fig. 7. Kotoite grains showing »Pavement-structure« in calcite. Plain light, x100.

In the places where ludwigite is in contanct with marble, in the rounded calcite granules of the marble a mineral harder than calcite having a nearly similar, or higher refraction, could be observed (Photo 7). The size of the granules does not exceed 1 mm. Owing to the extremely small amount their magnesium and boron content could only be qualitatively demonstrated. A thin section from a sample of szájbélyit from Rézbánya (Baita, Rumania) was made in view of the fact that *T. Watanabe* [1939] the discoverer of kotoite found the kotoite beside the szájbélyit of Rézbánya. In this section kotoite could also be detected and on comparing it with the mineral from Vaskő it could unequivocally be established that it is kotoite. Unfortunately, more thorough investigations could not be carried out because of the rare borate minerals from the Banat there was not enough substance available.

On the action of hydrothermal solutions ludwigite and kotoite decompose into szájbélyite. The fine needles, and needle aggregates of szájbélyit — as has already been established by *M. Lőw* [1911] — occur fairly plentifully in the hollows of ludwigite subsequently filled up by calcite. They can also be found as inclusions in the calcite granules of the marble in contact with ludwigite. The needles showing a very strong birefringence are arranged in radial aggregates.

Among the sulphides pyrite occurs far more rarely. I could only once observe its crystalline band along the ludwigite fibres. The sphalerite replaces both magnetite and ludwigite. Judging from its resin yellow internal reflexes it has a low Fe content. Calcite is the youngest primary mineral of the paragenesis. Its granules, always showing polysynthetic twin lamellae, contain the crystals and granules of the minerals mentioned hitherto, occur as inclusions (Photo 8). The ludwigite

Fig. 8. Polysynthetic twinned calcite with magnetite and ludwigite inclusions. Crossed nicols, x50.

alters into limonite. On the sections prepared from the altered samples splendid orange yellow coloured internal reflexes can be seen, the substance is pseudomorph after ludwigite, the thicker fibres of this mineral pass intactly through the parts already limonitised showing a ludwigite structure. As mentioned the breakdown of the thicker ludwigite fibres begins in the separation-directions perpendicular to the crystallographical axis *c*, thus sometimes dividing the ludwigite fibres into 3—4 parts (see Photo 1).

The dominating members of the interesting and rare mineral-associations are:

magnetite, ludwigite associated rock: contact marble, serpentine

Minerals of pneumatolytic origin only appearing in traces:

franklinite, kotoite.

Ores of hydrothermal origin only appearing in traces:

pyrite, sphalerite.

Mineral formed secondarily on the action of hydrothermal solutions:

szájbélyit.

Hydrothermal accessory mineral:

calcite.

Secondary weathering product:

limonite.

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CONTRIBUTIONS TO THE JAMESONITE PROBLEM

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Jamesonite is not a rare mineral. In spite of this there are questions to be elucidated concerning its composition, formula, ore microscopical properties as well as its relation to plumosite. Similarly the question of parajamesonite of Kisbánya (Herja) must also be cleared. Our task is to answer these questions.

INTRODUCTION

Among the lead-antimony sulphides jamesonite is relatively the most frequent mineral. In some localities e. g. at Zlatá-Ida (Aranyida, Czechoslovakia) it was the dominating ore of the veins. In spite of this, this interesting mineral is not well known.

Its formula was until recently assumed to be $Pb_2Sb_2S_5$ *[Eskola 1949, Köhler, 1949]*, although on the basis of analyses already in 1910 the formula $Pb_4FeSb_6S_{14}$ was proposed by *Schaller*. To day this is the generally accepted formula of jamesonite. At the same time in his work issued in 1953, *Machatschki* proposed the formula $Pb_4(SbFe)_7S_{14}$, whereas *Kostov* in his book published in 1957 uses the formula $Pb_4Sb_6S_{13}$.

The use of the name plumosite is not quite satisfactory and unequivocal. Usually the fine threaded cottonwool-feltlike jamesonite, occasionally boulangerite is denoted by this name, but according to *Ramdohr* and *Betechtin* the plumosite or »Federerz« is extremely fine threaded iron-free jamesonite. Among the occurrences of this iron-

free plumosite Baia Sprie (Felsőbánya, Rumánia) is mentioned by Ramdohr.

Having personally collected material from the best jamesonite occurrences within the Carpathians I and my coworkers began to examine this ore.

The chemical examinations were carried out by *Gy. Grasselly*, the roentgenographical ones by *K. Paděra*.

RESULTS OF THE MINERALOGICAL AND CHEMICAL EXAMINATIONS

Within the Carpathians the jamesonite appears genetically in two different ways. At Zlatá-Ida epithermal silver-bearing jamesonite veins connected to Variscian granites are known. The youngest sulphide ore of the vein is the dominating jamesonite.

The jamesonite in mineralized veins connected to Tertiary subvolcanic rocks can be found in the following places:

in mesothermal veins in Nagybörzsöny (Hungary) and Herja (Rumania), among the minerals of the mesothermal metasomatic ore body of Rodna (Rumania),

in epithermal silver-bearing zinc-lead veins of Gyöngyösoroszi (Hungary), Baňská Štiavnica (Selmecbánya, Czechoslovakia), Kereszthegy near Baia Mare (Nagybánya, Rumania), Baia Sprie (Felsőbánya, Rumania), Capnic (Kapnikbánya, Rumania),

in epithermal gold-silver veins of Valea Borcutului (Borpatak, Rumania), Săcărâmb (Nagyág, Rumania).

In this localities jamesonite occurs in economically insignificant quantities mainly as very fine-threaded plumosite.

Zlatá Ida (Aranyida, Czecho-Slovakia).

Jamesonite is the dominating ore of the epithermal veins genetically connected to the Variscian granite. The dark grey ore with silky metal lustre is a dense interwoven texture of thinner or thicker jamesonite threads reaching several cm in length. The ore replaces older idiomorphic pyrite occurring in far smaller amounts further sphalerite rich in chalcopyrite inclusions as well as freibergite and idiomorphic arsenopyrite. (Photos 1, 2, 3). Galena could not even be found in traces.

Among the older sulphide ores in sphalerite and freibergite gold grains with a diameter of 20—30 μ could be observed. Thus the slight gold content of the jamesonite originates from the older ores replaced by it. A part of its silver content certainly is derived from freibergite.

The quartz and siderite present in considerable quantities in the veins are mostly older than jamesonite, however, younger siderite replacing jamesonite could also be observed in polished ore sections.

In polished ore sections on the columnar-fibrous crystals of jamesonite sometimes a dense striation running parallel to the *c* crytallographical axis showing a fine threaded structure may be observed. Its

reflection pleochroism is not striking. The characteristic greenish tint is well visible. Beside jamesonite the freibergite is darker, its colour is brownish with a mildly pinkish shade. In oil immersion the difference in colour is more intensive. Arsenopyrite is lighter and has a white colour with a pronounced yellowish tint.

Under crossed nicols the anisotropy effects are strong, the colours (yellow — greenish yellow — slate blue) depend upon the orientation, however, they are less intensive than the colour shown by antimonite and berthierite. As is mentioned by *Ramdohr* the extinction is parallel

Fig. 1. Jamesonite (white) replacing sphalerite (grey), pyrite (gray with rough surface), freibergite (light grey) and arsenopyrite (white with strong relief). Plain light, x50.

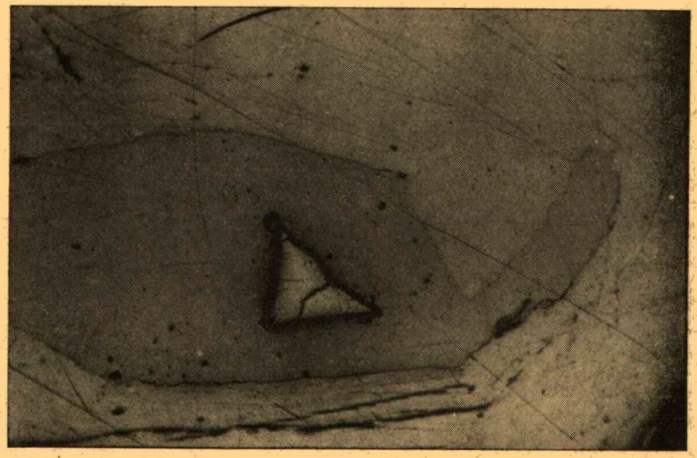

Fig. 2. Jamesonite replacing freibergite (grey). In the freibergite arsenopyrite as inclusion. Plain light, x50.

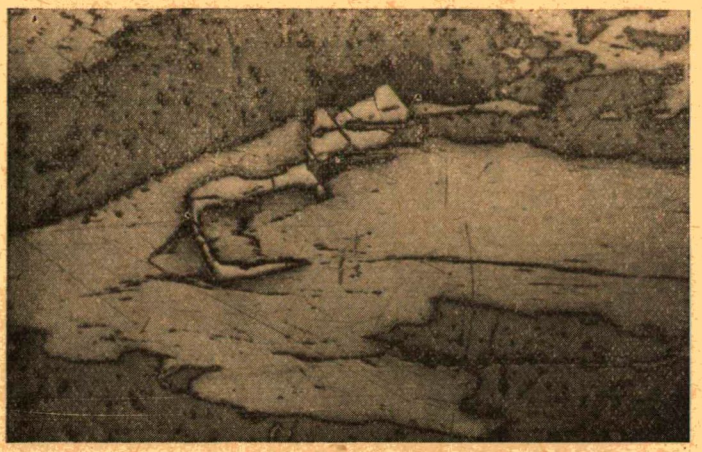

Fig. 3. Coarsely threaded jamesonite replacing arsenopyrite and sphalerite (grey). Plain light, x50.

but it will not be totally extinguished, the darkest colour is dark brownish grey. Among the wide jamesonite threads twin crystals can only rarely be observed. In contrast to *Murdoch*'s opinion it could be stated that besides ccHNO_3 also ccHCl attacks the jamesonite ore sections under development of H_2S. A dull slightly brownish grey patch forms and the crystal disintegrates into masses of fine threads running parallel to the *c* crystallographical axis. (Photo 4). Under crossed nicols the intensity of colours decreases considerably, the etched part shows a silky lustre.

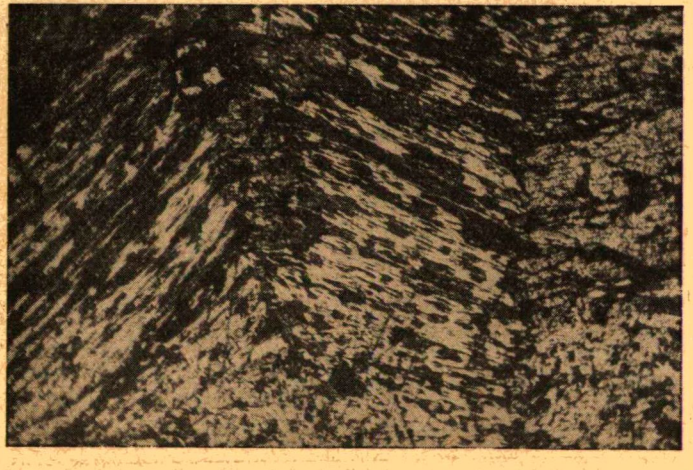

Fig. 4. Translation-twin lamellae of jamesonite developed by etching with HCl cc. Plain light, x100.

The columnar crystal aggregate built up from a mass of fine threads running parallel to the c crystallographical axis undergoes a translation under a tectonical load (Photo 5). Columns not etched often show a spindle-like translation phenomenon. The crystals and granules of the older, more rigid, ores are surrounded and replaced by jamesonite. The

Fig. 5. Translation phenomenon in jamesonite. Crossed nicols, x50.

jamesonite crystal aggregates intruding into small cavities, filled up subsequently by quartz or siderite, are branched. It can be well observed that the branched jamesonite fibres have mostly no terminal ends, they disintegrate into finer threads. Sometimes, however, the crystals are covered by curved platelets of a prism of fourth order. Of the jamesonite

Fig. 6. Twins of jamesonite with terminal planes embedded in quartz. x300.

crystals embedded in quartz twin crystals according to 100 can also be observed (Photo 6). The result of chemical analysis of jamesonite from Zlatá Ida is the following:

	1
Sb	27,95 %
Pb	31,69
Fe	2,13
Cu	0,08
Zn	13,30
S	23,80
SiO_2	1,10
	100,05 %

(Analyst: Mrs. E. Klivényi)

Samples for investigation were available from mesothermal pyrrhotite-sphalerite-galena veins connected to pyroxen-andesite, of Herja and mesothermal metasomatic pyrrhotite-sphalerite-galena-pyrite veins, connected to andesite, of Rodna. In its Tertiary subvolcanic occurrences the jamesonite occurs everywhere in two different ways: partly as thin covering layer consisting of dense aggregate of crystal-needles at the border of galena crystal groups replacing the galena, partly as extremely fine threaded loose plumosite forming cottonwool-like aggregates. The occurrence is economically insignificant everywhere.

Herja (Kisbánya, Rumania)

At Herja the jamesonite occurred very plentifully at the upper levels of the Salán vein. The galena-replacing jamesonite intruding frostwork-like into the galena is very frequent. (Photos 7, 8, 9). The replacement takes place starting from the boundaries of the coarsely crystallized galena aggregates of the cavity filling consisting in bulk of pyrrhotite-

Fig. 7. Jamesonite replacing frostwork-like galena. Crossed nicols, x50.

Fig. 8. Twinned jamesonite replacing frostwork-like galena. x100.

Fig. 9. Jamesonite replacing galena. Crossed nicols, x100.

sphalerite (14,69 per cent Fe). Beside the jamesonite, semseyite can be often found as sulphosalt, replacing galena. The tabular crystals or fan-like crystal aggregates of semseyite are replaced by the younger jamesonite (Photo 10). Jamesonite replacing semseyite intrudes as thin columnar crystals into it. The columnar crystals are often twins (Photo 11).

Fig. 10. Fine threaded jamesonite replacing tabular semseyite crystals. Crossed nicols, x100.

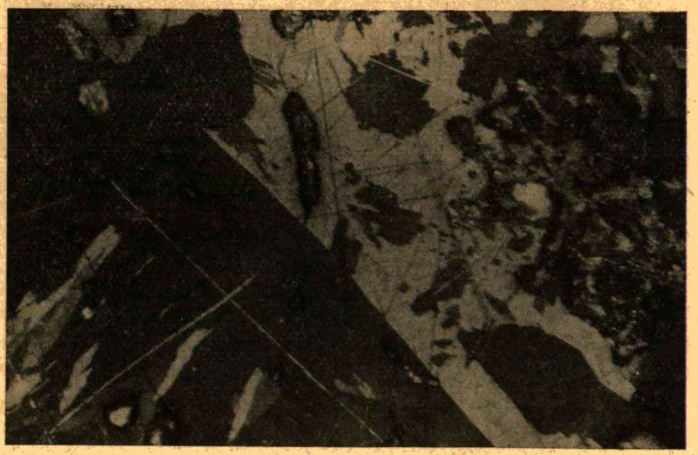

Fig. 11. Twin lamellated jamesonite replacing semseyite. Crossed nicols, x50.

The jamesonite layer consisting of dense aggregate of thin columnar crystals may be several mm in thickness. If the crystal aggregates of this layer intrude into the small cavities of the ore, they become extremely fine threaded feltlike mass. This feltlike plumosite covers the a few mm long, flat columnar jamesonite crystals of corroded surface, overgrown on the surface of the dense jamesonite coating (analysis 2).

These crystals — like the jamesonite forming their basis — are built up from frostwork-like interwoven crystal aggregates, thus they are no monocrystals. The occurrence and habit of these crystals agree with those of the parajamesonite crystals investigated by *Zsivny* and *Náray-Szabó*. The result of the chemical analysis of the dark grey plumosite coating covering these crystals and galena can be seen in analysis 3.

In the hollows of the ores of the Salán vein — particularly on level IV — the cottonwool-like soft texture consisting of interwoven aggre-

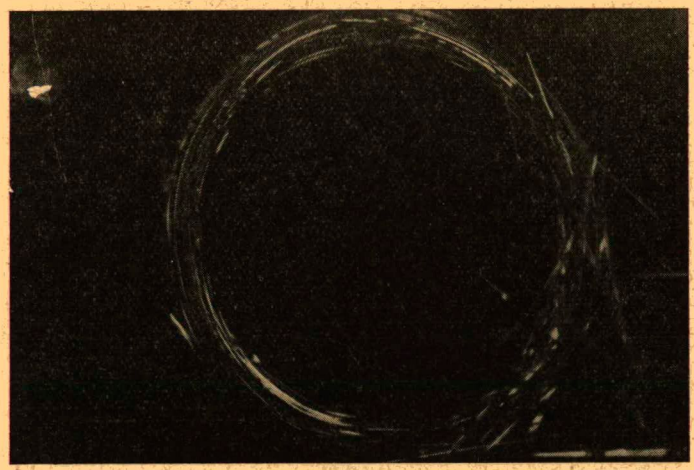

Fig. 12. Strongly curved bundles of jamesonite (plumosite) threads. x50.

gates of very fine needles of plumosite can often be found. From some hollows handfuls of fine threaded plumosite can be picked out. The threads are often curved, some threads and crystal aggregates built up from these threads even often form a circle (Photo 12). Overgrown on fine plumosite threads extremely minute ideally developed hexahedrons of pyrite, pointing to low formation temperature, can often be found. These pyrite crystals, which owing to their tinyness cannot be removed, cause the strikingly high iron content in the results of analyses of jamesonite specimens.

The semseyite, jamesonite as well as the relatively rare fizelyite formed at the cost of galena. The surface of the crystal aggregates of galena from Herja is very often corroded, the edges of the crystals are rounded and the surface of the crystallized specimens is often corroded, »parquetlike«. In the loose cottonwool-like mass of plumosite intergrown one can find the short columnar crystals of quartz, the $-^1/_2R$ rombohedrons of calcite attaining 1 cm in size as well as the globular crystal aggregates of siderite and dolomite stained grey or black by mass of plumosite inclusions (Photo 13). In the relatively rare crystals of gypsum and vivianite, plumosite also occurs plentifully as inclusions.

The composition of the loose cottonwool-like plumosite is given in analysis 4.

Fig. 13. Jamesonite (plumosite) inclusions in black calcite. Plain light, x100.

Analyses of jamesonite (plumosite) specimens from Herja:

	2	3	4
Pb	38,18 %	35,92 %	37,65 %
Sb	34,78	31,72	35,45
Fe	4,38	7,57	5,22
S	22,15	23,20	21,84
SiO_2	0,28	1,61	0,23
	99,77 %	100,02 %	100,39 %

Rodna (Óradna, Rumania)

In the lodes containing galena in Rodna the jamesonite occured everywhere. Here also the thin coating formed by frostwork-like crystal aggregates of jamesonite replacing galena as well as the plumosite masses composing loose cottonwool-like aggregates in ore hollows may be found. The plumosite occurs particularly in large amounts in the bedrock of the »Nándor« pyrite stock. In the hollows of the stock composed of pyrite—black sphalerite (14,27 per cent Fe), less galena and chalcopyrite, the cottonwool-like plumosite occurred frequently. Here also like at Herja the galena everywhere shows traces of dissolution, its crystals are rounded.

The dolomite crystals containing plumosite inclusions are stained grey and black by this mineral.

The result of the analysis of the fine threaded cottonwool-like plumosite from Rodna:

	5
Pb	37,52 %
Sb	33,76
Fe	4,94
S	21,98
SiO_2	1,71
	99,91 %

Baia Mare (Nagybánya, Rumania), Valea Borcutului (Borpatak, Rumania), Baia Sprie (Felsőbánya, Rumania)

The mode of the occurrence of the jamesonite at Baia Mare, in the mine of Kereszthegy, at Valea Borcutului and Baia Sprie is identical with the occurrence of Herja. In these localities too the jamesonite (plumosite) filling up the cavities as extremely fine threaded cottonwool-like aggregates as well as the galena replacing frostwork-like crystal aggregates can be found.

The mass of the plumosite inclusions stains black the calcite crystals at Baia Mare, whereas at Valea Borcutului and Baia Sprie beautiful baryte crystals and crystal groups can be found stained partly or completely black.

The well developed andorite crystals occurred at Baia Sprie in mine »Gyulaköze« in 1925 were embedded in plumosite.

The result of the analysis of fine threaded plumosite from Baia Sprie carried out earlier by *J. Loczka* is as follows:

	6
Pb	39,38 %
Sb	35,80
Fe	2,65
Mn	0,12
Zn	0,10
S	21,59
SiO_2	0,50
	100,14 %

Taking into account the analysis reported in the present paper as well as a few earlier results, the question was dealt with which of the formulas mentioned in the introduction can be accepted as the most probable formula for jamesonite.

In Table 1 the data of the analyses reported as well as those of the analyses according to *Loczka* and *Náray-Szabó — Zsivny — Zombory* are summarized, recalculating to 100 per cent the Pb, Sb, Fe, S content of the samples after subtracting in a suitable form the Cu, Zn, Mn, SiO_2 content present as impurities.

In the Table 1 also the formulas are denoted which are calculable from these data.

Table 1.

Specimen and No. of the analysis	Locality	Composition recalculated to 100 per cent		Mol. quotient		Formel
Jamesonite (1)	Zlatá Ida	Pb	40,17 %	0,1939	4,15	4PbS. FeS. $3Sb_2S_3$
		Sb	35,43	0,2910	6,23	
		Fe	2,61	0,0467	1	
		S	21,79	0,6797	14,55	
»Parajamesonite« (2)	Herja	Pb	38,38	0,1852	2,35	2PbS. FeS. $2Sb_2S_3$
		Sb	34,96	0,2871	3,64	
		Fe	4,40	0,0788	1	
		S	22,26	0,6943	8,81	
Plumosite (3)	Herja	Pb	36,51	0,1762	1,27	4PbS. 3FeS. $3Sb_2S_3$ x3
		Sb	32,23	0,2647	1,92	
		Fe	7,69	0,1377	1	
		S	23,57	0,7352	5,33	
Plumosite (4)	Herja	Pb	37,59	0,1814	1,94	4PbS. 2FeS. $3Sb_2S_3$ x2
		Sb	35,40	0,2907	3,11	
		Fe	5,21	0,0933	1	
		S	21,80	0,6800	7,28	
Plumosite (5)	Rodna	Pb	38,21	0,1844	2,04	4PbS. 2FeS. $3Sb_2S_3$ x2
		Sb	34,38	0,2824	3,13	
		Fe	5,03	0,0901	1	
		S	22,38	0,6981	7,75	
Plumosite (6)	Baia Sprie	Pb	39,67	0,1914	4,02	4PbS. FeS. $3Sb_2S_3$
		Sb	36,06	0,2962	6,23	
		Fe	2,65	0,0475	1	
		S	21,62	0,6744	14,19	
»Parajamesonite«	Herja	Pb	40,01	0,1931	3,59	7PbS. 2FeS. $5Sb_2S_3$ x2
		Sb	34,92	0,2868	5,34	
		Fe	3,00	0,0537	1	
		S	22,07	0,6884	12,81	

The formulas which can be calculated from the above data are not unequivocal, especially not in the case of the two parajamesonite specimens, whereas in that of the other samples besides the PbS/Sb_2S_3 ratio of constant character, the FeS content fluctuates. It is to be noted that on the polished ore sections prepared from the samples analyzed (analysis 2) besides the jamesonite and parajamesonite also another lead-antimony sulphosalt (semseyite) can also be found, thus the fluctuation in the formula is probably due to the presence of other similar sulphosalts.

The formulas were also calculated on the basis of earlier jamesonite analyses considering the iron content as belonging to the jamesonite

molecule. Hence on the basis of the data of analyses XXIV, XXXIV, XXXVII from Hintze's Handbuch der Mineralogie (Bd. I. 1. p. 1032) as well as on that of the analyses 5., and 7., from Dana's System of Mineralogy (Vol. I. p. 453), the following formulas for jamesonite can be calculated:

$4PbS.\ 2FeS.\ 3Sb_2S_3$
$2PbS.\ FeS.\ Sb_2S_3$
$4PbS.\ FeS.\ 3Sb_2S_3$
$4PbS.\ FeS.\ 3Sb_2S_3$
$3PbS.\ FeS.\ 2Sb_2S_3$

Thus the formula of the jamesonite varies also on the basis of the data of earlier analyses — especially changes the iron content — like the formula calculated from the recent data.

At the same time, however, if the iron content is not considered to belong to the jamesonite and only the Pb, Sb, S content is recalculated to 100 per cent after subtracting the iron content and the corresponding S content with the other impurities, in the case of all samples the formula of $4PbS.\ 3Sb_2S_3$ is obtained, corresponding to the formula $Pb_4Sb_6S_{13}$ given by *Kostov*.

As, however, we have never found iron-free jamesonite samples it may be assumed that the formula $4PbS.\ FeS.\ 3Sb_2S_3$ suggested already also by *Schaller* and confirmed by other authors can be considered as the formula of jamesonite. It should be remarked — and this explains the more or less fluctuating iron content — that a part of iron indeed often does not belong to the jamesonite molecule but it is present as impurity. This is also confirmed by the fact that on the fine threads of numerous plumosite specimens overgrown pyrite crystals in microscopical size could be observed and the sample to be analyzed could not be separated from them. In the case of such finely threaded substances as plumosite an entanglement with impurities, which can only difficultly or alltogether not be removed, must be taken into account.

THE RESULTS OF THE X-RAY EXAMINATIONS

The possible identity of parajamesonite with jamesonite

Zsivny and *I. Náray-Szabó* [1947] described from the locality of Herja (Kisbánya, Rumania) a new mineral which they called parajamesonite. Jamesonite and parajamesonite having the same chemical formula ought only to differ markedly by the powder roentgenogram. We were therefore very surprised to observe that the x-ray diagrams of the samples from the original locality of Herja (Kisbánya) were identical with those of the jamesonite.

At the x-ray investigations two samples of parajamesonite from Kisbánya (Herja) were available. The Debye-Scherrer method was applied. The conditions of the exposure were as follows: Cu anticathode, Ni filter, chamber diameter 57,4 mm.

The intensities of the interference lines were estimated in a scale of ten degrees.

Five diffraction patterns were made which always showed the identity of the dominant component of the specimens with jamesonite. To eliminate the possiblity of the modification-change due to the grinding of the samples to be investigated, three exposures were made from the needlelike crystals of parajamesonite with a normal metallic lustre, ground during various periods. The different grinding time i. e. not quite a minute, about 10 minutes and more than an hour, did not influence the *d*-values and the intensities of the interference lines. However, the exposure of the parajamesonite powder not ground for quite a minute furnished two different interference lines. The stronger interferences showed uninterrupted arches and belong to jamesonite. The weaker interference lines were composed of single divided dots. They often coincided with the preceeding interference lines and than their presence was revealed by the appearance of the single dots on the normal diffusion arch. Their *d*-values in Å units 3,10—1,91—1,64—1,245—1,112——1,046 : 0,958 and 0,918 correspond to the strong interferences of sphalerite.

The presence of some of the strongest lines of sphalerite could also be observed at the diffraction patterns of parajamesonite powder ground during a longer period. In this case, however, it was not possible to distinguish the interferences of sphalerite from the coinciding lines of jamesonite, as the latter were far more intensive against the interference lines of sphalerite. The identification of the subordinate component i. e. of sphalerite in the samples investigated rendered possible only by the diffraction pattern of the powder ground for a short time. The fourth x-ray diffraction pattern is made from the black powder with a non metallic lustre which coats the not well developed needlelike crystals of parajamesonite. The evaluation of the diffraction pattern and the comparison with the results obtained by *L. G. Berry* [1940] for jamesonite are comprised in Table 2. They proved that the black powder is identical with jamesonite.

Table 2.

Jamesonite, Kisbánya (Herja) Cu/Ni			Jamesonite according to L. G. Berry	
No of the lines	I	d		d
1	4	4,04 Å	s	4,03 kX
2	6	3,77	s	3,76
3	1	3,69		
4	2	3,55		
5	10	3,40	vvs	3,42
6	2	3,30		
7			vvw	3,20
8	3	3,13	s	3,11
9	5	3,08		
10	3	2,94	w	2,95

Jamesonite, Kisbánya (Heria) Cu/Ni			Jamesonite according to L. G. Berry	
No. of the lines	I	d	I	d
11	9	2,81	s	2,81
12	8	2,71	vs	2,72
13	2	2,62		
14	3	2,35	vw	2,37
15	5	2,29	s	2,31
16	5	2,24	s	2,25
17	3	2,18	vvw	2,17
18	3	2,10		
19	6	2,04	vs	2,05
20	7	2,02	vs	2,04
21	2	1,95	vw	1,967
22	4	1,90	m	1,905
23	3	1,87	w	1,882
24	7	1,83	s	1,826
25	1	1,78		
26	2	1,76	vvw	1,760
27	4	1,71	m	1,717
28	1	1,65		
29	2	1,61	vvw	1,616
30	2	1,57	vvw	1,581
31	3	1,52	m	1,528
32	1	1,480		
33	4	1,449	vw	1,455
34	3	1,414	vw	1,415
35	2	1,382		
36	3	1,369	m	1,361
37			vw	1,347
38	2	1,313		
39	2	1,287	w	1,283
40	2	1,261		
41	2	1,234		
42	3	1,212	m	1,208
43	1	1,184		
44	4d	1,165	m	1,169
45	5d	1,118		
46	2	1,082		
47	1	1,064		
48	1	1,048		
49	1	0,993		
50	2	0,976		

The diffraction pattern of parajamesonite obtained by *V. Zsivny* and *I. Náray-Szabó* [1947] differs completely from our results obtained for jamesonite. The recalculated *d*-values of the results as well as the intensity of the single interferences concerning the parajamesonite obtained by the authors mentioned above are illustrated in Table 3.

Table 3.

Parajamesonite
Kisbánya (Herja)

No of the lines	I	d
1	3	4,66 Å
2	8—9	4,21
3	5	3,78
4	1	3,29
5	1	3,00
6	1	2,73
7	1	2,65
8	2	2,49
9	2	2,23
10	2	2,02
11	1	1,92
12	1	1,84

The fifth x-ray pattern was obtained from a second specimen of parajamesonite. The substance to be investigated was separated from the needle-like incompletely developed crystals with a metallic lustre.

The roentgenographical identification proved that the specimens from the Herja locality or at least most of them, considered in the collections as parajamesonites, are actually jamesonites. Whether or not parajamesonite is identical with jamesonite or if truly an independent mineral is involved, can only be decided by further investigation of parajamesonite specimens from the original locality. The rareness and inaccessibility of the further specimens of parajamesonite rendered it impossible for the authors to solve the problem decisively.

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DATA ON THE FORMATION OF THE KAOLIN OF THE TOKAJ-MOUNTAINS

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SUMMARY

In the southern part of the Tokaj-Mountains the kaolin and bentonite deposits are embedded in rhyolite-tuff-rhyolite of Tortonian-Sarmatian age. These have formed partly due to endogenic and partly exogenic process from orthohypomagmatites, pyroclastics and tuffits. While, in the case of the kaolinization of Végardó — Sárospatak and Mád may also be ranged here — the greater part of the basic rock is tuffit, at Bodrogszegi true pyroclasticum occurs. Here the action of the hydrothermae was the slightest, lasted the shortest time and the exogenic processes exerted their effect to the greatest extent.

Usually the formation-sequence of the mineral associations of the different deposits can be traced well. The changes of the pH can be well observed also in the occurrence of some of the characteristic minerals. The formation — sequence of the minerals corresponds to the observed one and it may also be characterised by the decreasing compound potentials.

The formation of the clay minerals has since a long time been of interest to investigators as well as to the parties concerned in ceramic and porcelain industry. As clay minerals from various ore deposits were investigated by different methods, the opinions concerning the formation differed also to a great extent. Besides the circumstances of the formation the starting rock plays also an important part. It is known that generally the feldspars and the glassy matrix of the tuffs and the acidic and to a smaller extent that of the intermediary and alkaline rocks compose the basis for the formation of the clay mineral. Hence, the clay minerals occur as secondary minerals. Both in the case of kaolinization and bentonitization regarding their formation the tracing of the weathering of the feldspars is an essential point of view.

The more important data of the literature referring to the kaolinization

On the basis of the examinations of *Correns* and *Engelhardt* [1939/40] the components of the feldspar dissolve as ions. The velocity of the weathering process increases with the decreasing grain-size and increasing temperature. Before the kaolinazition the lattice of the feldspar decomposes.

According to *Schiebold* [1931] at the weathering of the feldspars the alkalies on the surface of the lattice plane are in a very exposed situation, therefore, they dissolve easily. It has also been established that the presence of iron does not interfere with the kaolinization even Mn, Ca, Na, K only interfere if their total quantity related to that of Al_2O_3 does not exceed 0,02 : 1.

Frederickson [1951] states that at the weathering of the silicates in the first place at that of the feldspars the most exterior unbound oxygen ions of the silicate lattice attract one of the two free positive charges of the H_2O molecules, whereas the valency excess of the Al tetrahedrons is bound by the other free positive charge which in the feldspar was originally bound by K or Na.

Andreatta [1949] found that if the quantity of the water increases suddenly the minerals with a small compound potential dissolve first, whereas, of the minerals with a high compound potential, the elements of low ionic potential dissolve. Owing to this the weathering of the feldspars begins with the dissolution of the alkalies.

At many occurrence of hydrothermal clay minerals a zonal arrangement of them could be observed. According to *Sales* and *Meyers* [1948] the formation of the clay minerals is essentially simultaneous with the deposition of the ores, it is syngenetic and the development of the different zones may be considered as a reaction zone. From the original rock toward the hydrothermal vein the amount of Mg, Ca, Na and Si changes considerably.

Grim [1953] established that kaolinit may form both from acid and intermediary rocks, if at the weathering of the feldspars the alkalies and alkaline-earth metals disappear as quickly as they are released from the feldspar. The presence of Ca in the environment is not favourable for the formation of kaolinite.

Bates and Comer [1955] examined the weathering of the feldspars electron microscopically and found that on the cleavage plane of the microclin sometimes hexagons, which are probably kaolinites, may be detected.

At the weathering in addition to the sudden change of the water-amount, the pressure and temperature also change as compared to the conditions at the formation of the magmatic rocks, as it has been described in detail by *Szádeczky-Kaross* [1958—59]. Whilst the decrease of the pressure attempts to diminish the coordination number, the decrease of the temperature endeavours to elevate it. On the surface, the latter will play a considerable role. This change is particularly striking in the case of Al.

Stringham [1952] has summarised by the elaboration of the data of the literature the possibilities for the formation of clay minerals under hydrothermal conditions.

In order to elucidate the formation of the clay minerals many synthetic experiments have been carried out. Of these the experiments of *Noll, Gruner* and *Folk* are the most interesting.

The synthetic examinations of *Noll* [1935—1936] point to the fact that really speaking the formation of kaolin is a result of a very simple reaction, which takes place between Al_2O_3 and SiO_2 in the presence of water, i. e. kaolinization may occur both under hydrothermal and epigen conditions.

According to *Gruner* [1944] the conditions for the formation of the single clay minerals are determined by the concentration of the K ion as well as by the ratio of the Al_2O_3 and SiO_2.

Folk [1947] also states that the ratio of the Si: Al and the K content determines which mineral forms synthetically if the temperature and the pH are suitable.

THE CONDITIONS OF THE OCCURRENCE

Already since a long time investigators have dealt with the metamagmatites of the Tokaj-Mountains. While earlier principally the kaolinizaton of the extensive rhyolite-tuffs was taken into account recent investigations also the bentonitization of the rhyolite-tuffs have revealed. These recent investigations have shown that at the southern part of the Tokaj-Mountains there are four important kaolin occurrences: the kaolin deposit at Végardó, the occurrence in the northern vicinity of Bodrogszegi, the environment of the mine of Mount Bomboly in the eastern vicinity of Mád and the occurrence at Mount Megyer to the north of Sárospatak. Of these the occurrences of Végardó and Bodrogszegi will be dealt with in detail.

According to recent investigations the geological structure of the Tokaj Mountains is as follows. Tortonian rhyolite-tuffs with great gaps in the sedimentation settled onto Carboniferous and Triassic formations [*Pantó—Mrs. Széki—Fux,* 1959]. In the Sziget-Mountains of Zemplén situated in the vicinity, the same situation prevails [*Ferenczi,* 1943] Owing to the volcanic activity which started in the Tortonian a great deal of volcanic detritus has come to the surface. A part of these still fell into the shallow sea, which is also proved by the fossils found in them, in other places where the volcanic detritus accumulated in greater thickness true pyroclastic formed. After the andesite eruption succeeding it another rhyolite volcanic eruption followed in the Sarmatian producing both lava and tuff. Interesting data was obtained from a deep boring to the north of Sárospatak it opened up at a deepness of 225 meter, directly under the rhyolite tuff, Triassic limestone. According to the assumptions of *Vadász* and *Ferenczi* [1943] it composes a connecting link between the Sziget-Mountains at Zemplén and the Mountains of Szendrő-Rudabánya.

In the Tokaj-Mountains both the rhyolite tuffs and rhyolites of Tortonian and Sarmatian age are kaolinized and bentonitized. To separate them is mostly a very difficult task. Generally, however, the Tortonian formations are rather bentonitized and in the Sarmatian formations, more intensive kaolinization may only be found at the places where hydrothermal solutions exerted their action on these formations. The general kaolinization extending over the whole mountains is the result of an epigen effect.

Végardó

Frits [1955—1956] described the kaolin and bentonite deposit of Végardó. According to the fossils determined by *Schréter,* the kaolin and bentonite deposits opened up by drilling and underground mining are the alteration products of rhyolite tuff and rhyolite, respectively, of Sarmatian age. On the basis of present knowledge the kaolin deposit is the deepest. Over it there are three bentonite deposits interrupted by rhyolite tuff interdepositions. Occasionally the rhyolite tuffs and rhyolites are impregnated with pyrite. Pyrite appears in the form of very small crystals and its crystal shape is usually very simple.

The relatively fine granular composition of the kaolinous bentonitous deposit of Végardó is striking. The amount of the granules larger than 1 mm only rarely exceeds 10 per cent, mostly it is far smaller. Its greater part is rock fragments, angular and lobate quartz. The crystal fragments of quartz are never rubbed they have not been transported, occasionally their surface is corroded.

Fig. 1. Angular, lobated quartz fragment. Rhyolite tuff. Crossed nicols, x50.

The result of the mechanical analyses of some samples is shown in Fig. 2., accordingly the fraction not exceeding 0,25 mm usually ranges between 65—75 per cent. Although cases have occurred in which this fraction did not even attain 50 per cent, in these cases, howewer, it is shown by microscopical examinations the matrix is silicified to a smaller or greater extent distorting be original values of the grain-size.

In the case of several samples a fraction not exceeding 0,25 mm was cleaned. In almost 90 per cent of the cases the diameter of the grains of this fraction did not exceed 0,02 mm.

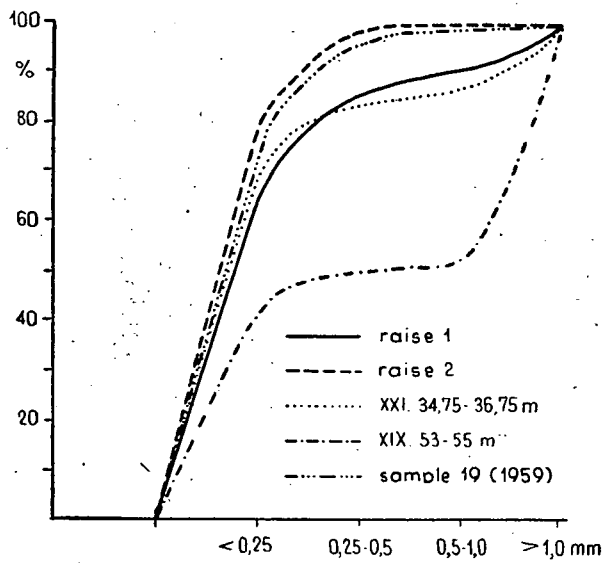

Fig. 2. Summary-curves of different kaolin samples from Végardó.

In the course of kaolinization an appreciable amount of silicic acid is released. Usually the released silicic acid separates as an opal. In the course of the weathering the silicic acid migrates towards the intact less decomposed rock. A good example for this is furnished by the occurrence in Végardó. At the raise of the subadit one finds in the lower part porous, pyritous in some places decomposed slightly kaolinized rhyolite-tuffs. Above the separation of the silicic acid appearing in a sharp line the tuff is strongly kaolinized it already belongs to the kaolin body. In this part the pyrite content is also minimal. The opaline part separating the two parts is 1—3 mm thick dark grey and sometimes permeated with pyrite veins. The pyrite veins run parallel to the opal zone and do not reach as far as the kaolin or the tuff part. Thus probably the pyrite contained in the opal is a syngenetic separation. The thinner and thicker opal veins can also be well detected under the microscope (Fig. 3.).

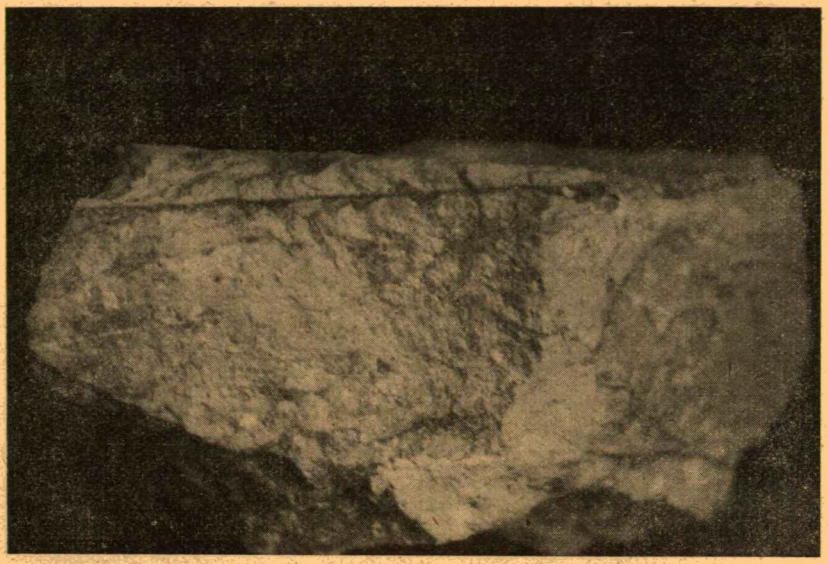

Fig. 3. Kaolinized and tuffaceuos part separated by an opal cord. $x^2/_3$

Another appearance of pyrite is also known when it appears in larger granules in the lower levels at the border of the kaolinized rhyolite tuff parts. On these chalcedony can always be found as one of the most recently separated minerals (Fig. 4.).

Fig. 4. Globular chalcedony on pyrite. x40

Kaolinite plays the most important role in the mineral composition of the kaolinized parts of Végardó. The DTA curve of the sample originating from the raise I of the A—73 levels has an ideal course. According to the examinations of *Földvári-Vogl—Koblenz* nakrite and dickite only appear in certain part in a subordinate amount and the

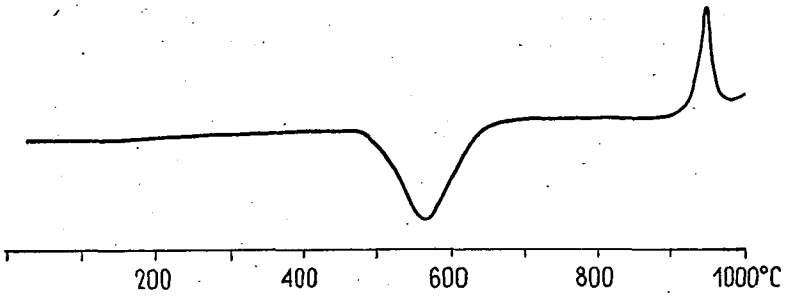

Fig. 5. The DTA curve of kaolinite from Végardó.

appearance is not connected with hydrothermae of high temperature. Their formation was more probably rendered possible through special conditions which are, however, as yet hardly known.

In some borings alunite also occurs. According to the chemical analyses the K_2O and Al_2O_3 content is high and the SiO_2 content is low. On the DTA curve besides the endothermic peaks characterising alunite the endothermic peak of the kaolinite also appears, thus between 500—600° C a double peak develops, the one appearing at lower temperature relates to alunite. For the sake of comparison the DTA curves of crystallized alunite of Beregovo as well as that of the mixture of this alunite with kaolinite from Végardó in a ratio of 1 : 1 were prepared. In the case of crystallized alunite at 570° C and 840° C

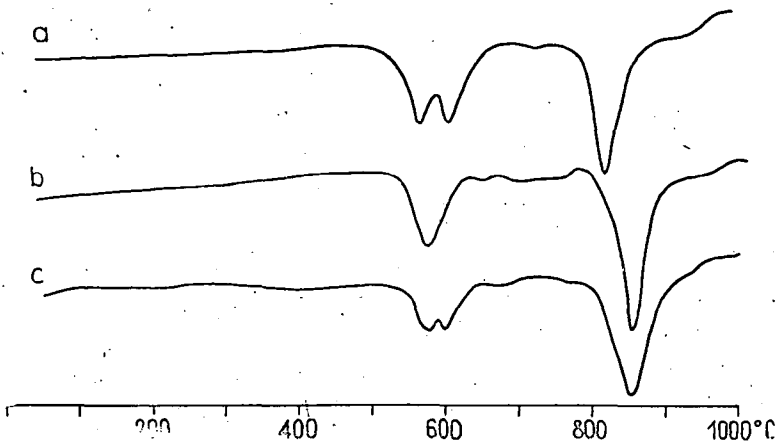

Fig. 6. a) Alunite with kaolin from Végardó, *b)* crystallized alunite from Beregovo, *c)* the DTA curve of a mixture of crystallized alunite from Beregovo and kaolinite from Végardó in a ratio of 1 : 1.

pronounced endotherm peaks appeare, at 620° C a smaller one, whereas at 750° C an exotherm peak can be found. On the DTA curve of the mixture mentioned above between 500 and 600° C the double peak developed and the endothermic peak appearing at lower temperature was coincided exactly with that of the alunite. At a little higher temperature the second endothermic peak appeared and disappeared the exothermic peak of 770° C.

According to *Kulp* and *Adler* [1950] the maximum of the first high endotherm peak appears around 550° C and the second around 810—850° C. Both denote a transformation of the lattice structure. At about 760° C a small exotherm peak appears pointing to the recrystallization of α Al_2O_3.

On examining the alunite-kaoline deposit at Beregovo, *Paluektova* observed [1957] on the DTA curves the first endotherm maximum at 590° C which means the disappearance of the structurally bound water and the second endothermic maximum at 845° C, when SO_3 group is released. At 765° she also observed a small exotherm peak.

In the case of the alunitous kaolin from Végardó the first endothermic peak also appeared at 600° C, the second one at 800° C. The exothermic peak only appeared in a blurred manner at no definite temperature and the exothermic peak of the kaolinite appearing between 900—1000° C, was also blurred.

The electron microscopical investigation also confirms the results of the DTA. The photos prepared from the suspension also give an

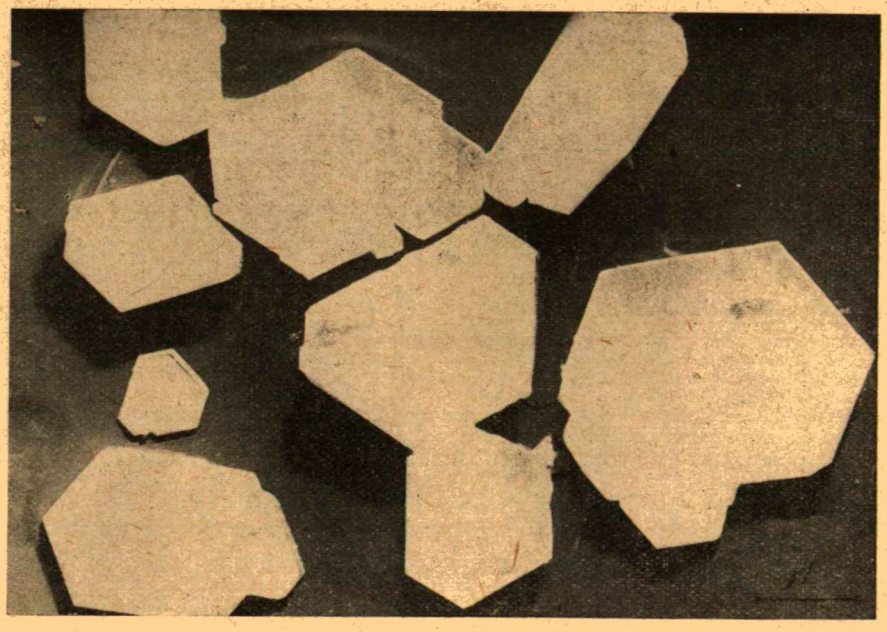

Fig. 7. Kaolinite crystals from Végardó.

ideal or slightly distorted picture of the kaolinite crystals. The size of the crystals is about 3 μ, whereas the thickness of the crystal plate calculated from the shadow is about 0,6—0,7 μ (Fig. 7.).

The photos are very interesting. The crystals are oriented arranged in such a manner as if the whole would be a single crystal. The edges are all parallel. The shadowings render well perceptible the pseudo-hexagonal plates of the kaolinite crystals. The size of the crystals varies, its measurable size lyes between 1—3 μ (Fig. 8.).

On the basis of the chemical composition of the kaolinized rocks and kaolins of Végardó the results of the analysis have been recalculated into elements which are summarized in the Table 1. The table also illustrates the trace elements.

Table 1.

	1	2	3	4	5	6	7
$Si^{4+} \cdot 10^2$	2171	2733	3368	3018	3332	3323	1485
$Al^{3+} \cdot 10^2$	1982	1436	941	1238	945	1054	1642
$Ti^{4+} \cdot 10^2$	10	20	29	29	27	17	17
$Fe^{3+} \cdot 10^2$	12	84	111	63	40	48	24
$Fe^{2+} \cdot 10^2$	—	1	7	—	tr	17	—
$Ca^{2+} \cdot 10^2$	9	14	20	23	25	20	31
$Mg^{2+} \cdot 10^2$	2	16	2	2	4	4	6
$Na^{1+} \cdot 10^2$	16	30	—	tr	tr	2	92
$K^{1+} \cdot 10^2$	24	30	6	tr	37	5	177
$P^{5+} \cdot 10^2$	13	7	—	13	8	—	—
$S^{6+} \cdot 10^2$	—	54	251	79	64	10	629
B	(tr)						(tr)
V							(tr)
Cr							(tr)
Co							(tr)
Ni							(tr)
Cu	tr						(tr)
Sn							(tr)
Pb	tr						(tr)
Mn	tr						(tr)?
As							(tr)
Si/Al	1,09	1,90	3,58	2,44	3,55	3,15	0,90
Al/Fe^{3+}	165,16	17,05	8,48	19,65	23,62	21,96	68,42
$K + Na \cdot 1000/Al$	20,18	41,73	6,38	0,81	39,15	6,64	163,82
K/Na	1,50	1,00	6,00	—	37,00	2,50	1,92

(tr)? — doubtful trace
(tr) — very weak line of spectrogram
tr — weak line of spectrogram

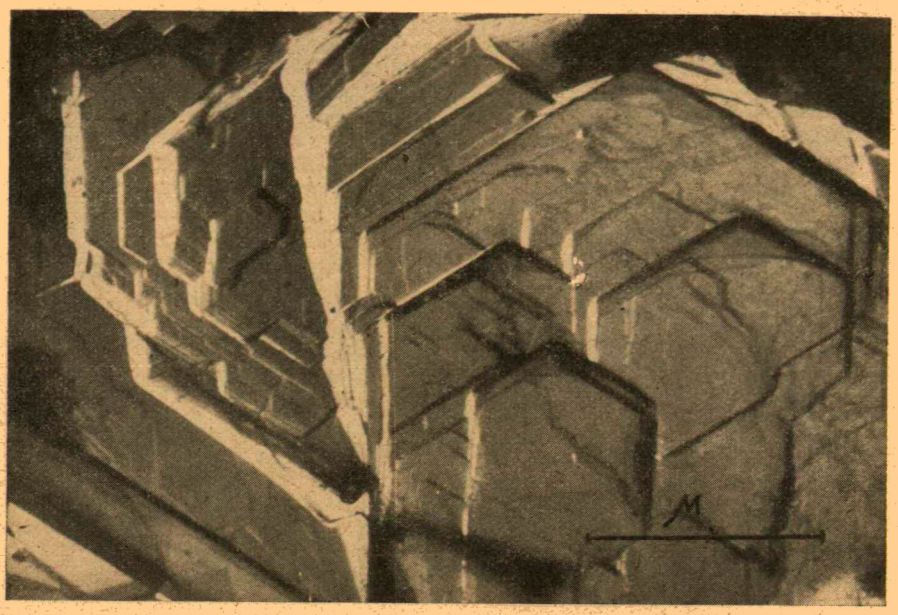

Fig. 8. Electron micrograph of kaolinite from Végardó prepared by the replica method.

Sampling places:

1. I. level, 1. raise 47,5—49,5 m,
2. three borings, the average of 59,03—76,43 meters,
3. boring No. IX, from 79,28—79,48 m, wtih much pyrite,
4. boring No. XIX. from 47—48 m,
5. boring No. XIX. from 48,5—51,0 m,
6. boring No. XIX. from 53,5—55,0 m,
7. boring No. XXI. from 34,75—36,78 m.

Of the results mainly the interpretation of the different quotients will be emphasised.

In the ideal kaolinite formula the Si/Al ratio is 1,04. Of the above values this is the most nearly approached by the substance obtained from the raise of level I which has also been proved by means of other examinations to be kaolin of a high quality. In the sample from boring XXI this value is only 0,9. This substance is, however, alunitized and the greater part of the Al is bound by this mineral. This is also de-

monstrated by the low Si value, thus a kaolin member containing more Al cannot be involved. In the other samples the Si/Al ratio is double or even triple, hence the substance is silicified which can also be observed under the microscope.

The Al/Fe^{3+} value shows the pureness of the kaolinite. The higher this number the more valuable the kaolin. A value about 20 still represents a good quality, decreasing values mean decreasing quality. Of the analyzed samples particularly the substance from the breaking out is very pure.

The $Ka + Na \cdot 1000/Al$ value shows really speaking the leaching factor. According to the examinations of *Laughlin* [1959] when the leaching increases and the weathering of the feldspars advances this value decreases also in the finer fractions. This is related to the fact that when the size of the granules decreases, the feldspar alters more easily, which has already been pointed out by *Correns* and *Engelhardt* [1939/40]. In the case of the above analysis this value ranges between wide limites showing the different extent of the kaolinization.

The base exchange possibilities influence somewhat the K/Na ratio, however, in the case of the kaolins it gives usually the ratio of the not weathered feldspars as in this case the cation exchange has a low value. In most cases, however, the decomposition of the feldspars is so great that the analyses either altogether do not contain alkalies, or only in a very insignificant percentage.

In the above analyses the FeO content is very minimal often it is lacking which proves that the weathering of the rock, the process of kaolinization took place under oxidative conditions.

The examinations of the minor elements show that the hydrothermae related to the hypovolcanites contain copper and lead in traces. A part of the copper and lead is bound to pyrite. As, however, these two elements only appear with a very weak spectogram line, their greater part can be found in the kaolinized parts, where these two components show a weak spectogram line. According to the examinations of *Laughlin* [1959] the V, Cr and Sn presumably substitute the iron in the octahedral layer. The Co and Ni give a very weak spectogram line. Ca and Mg appear in all cases with a strong spectogram line. Of the sedimentophil elements only B appears is very weak traces. It is interesing that Au, Ag, Bi, Zn, Zr, Be and Ge could not be detected. In the pyrite besides the Cu and Pb only As appeared with an averagely strong line, the Mn and Sb are questionable. Thus this pyrite is fairly pure and approaches the ideal composition.

The hydrothermae breaking up also at present are bound to fault lines. Their composition is fairly uniform. Hydrocarbonatous — sulphatous waters dominate in which Ca and Na are the dominating and Mg the subordinated cations. In small amounts iron may be found. The pH value is generally about 7. The thermal water break up from a depths of 70—90 m. Apparently the kaolinized-bentonitized zone plays the role of a water closing layer. At present their temperature does not exceed 30° C.

Table 2 shows the chemical composition of the hot waters.

Table 2.

	1		2		3		4		5	
	mg/l	equ. %	mg/l	equ. %	mg/l	equ. %	mg/l	equ. %	mg/l	equ. %
Na	219,6	41,99	202,5	38,48	218,0	41,73	360,2	65,93	341,5	66,95
Ca	194,3	42,64	200,3	43,68	196,7	43,21	99,0	20,85	97,4	21,91
Mg	42,1	15,23	47,7	17,14	41,6	15,06	31,1	10,79	24,8	9,19
Fe	—	—	3,5	0,55	—	—	16,0	2,42	12,0	1,94
ammonium	0,6	0,14	0,6	0,14	tr	—	tr	—	—	—
Cl	88,4	10,97	87,6	10,79	89,8	11,15	89,4	10,64	82,8	10,53
H_2CO_3	472,3	34,02	488,1	34,94	474,7	34,23	530,8	36,70	469,8	34,70
SO_4	600,8	55,01	596,3	54,26	596,0	54,62	599,2	52,66	583,4	54,77
NO_3	—		—		—		tr		—	
meta silicic aoid	19,5	—	28,6	—	28,0	—	—	—	5,2	—
free carbonic acid	—	—	138,6	—	129,3	—	156,4	—	81,1	—
total dissolved salt	1637,6		1793,8		1774,1		1882,1		1698,0	
alkalinity	7,74				7,78		8,70		7,70	
total hardness	36,90				37,12		21,03		19,33	
carbonate hardness	21,67				21,78		24,36		21,56	
O_2 consumption mg/l					1,6					

Places of water sampling:

1. from raise from subadit
2. boring 22. at the end of the subadit
3. boring IX/1. 88,09—88,59 m
4. boring XI. 76,0—77,9 m
5. boring XII. 82,1 m.

The hydrothermae are very uniform. This is also well illustrated by the diagram (Fig. 9.). Perhaps only the water breaking out from borings IX/1 and XI shows some difference namely the Ca and Mg is lower and the Na content higher. The anions, however, are present in the same amount. It is also characteristic of these waters that they contain a fairly appreciable amount of free carbonic acid.

At the investigation of the origin of the kaolin-bentonite deposit of Végardó it may be assumed that the rhyolitic volcanic fragments fell into the sea at the beginning which on the action of the sea water started to diagenese. The higher pH of the sea water, as well as the fact that the alkalies dissolved created a suitable physico-chemical environment for the beginning of the decomposition of the feldspars of these tuffs and tuffits, respectively, i. e. for the beginning of the leaching of the alkali content of the binding substance of the tuffs.

Later the hydrothermal solutions breaking out in connection with the hypomagmatits brought about the pyrite impregnation which can be seen in certain places in the rhyolite tuffs. At the weathering of the pyrite with plentiful solvent in an oxygen rich environment — these occur in the oxidation zone — an environment with a low pH developed on the action of sulphuric acid. The low pH value, the potassium

free and oxygen rich environment made the kaolinization possible. This was particularly favourable along the fault lines quite close to the hydrothermae. If there also occurred a part of tuff in which the feldspar did not decompose completely the movement of the solutions was fast enough to remove the dissolved alkalies and that finally kaolin should form.

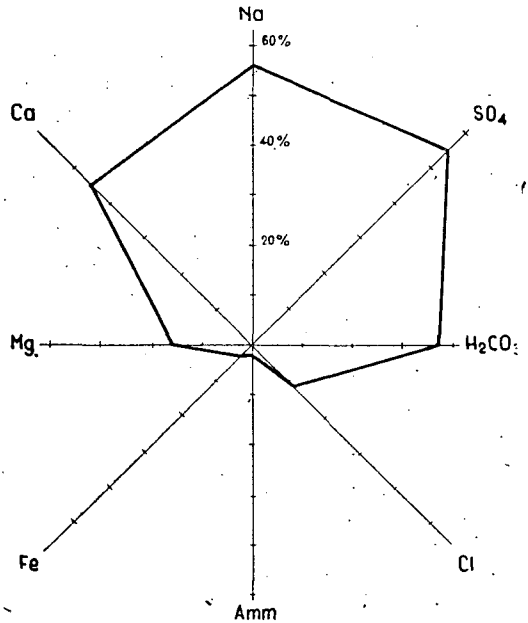

Fig. 9. The chemical composition of the hot water of Végardó.

Taking the above into account the formation of the minerals occurs probably in the following sequence. After the bentonitization of the early tuffits following the appearance of the hydrothermae, i. e. in a certain respect simultaneously with it, first kaolinite appeared associated in some places with alunite. The lacking of halloysite and allophane point to the fact that at the formation of the deposit epigen processes did not play an essential role. Although these form at a low pH value the temperature must have also been very low. With the advancing of the weathering of the residual feldspars and with the solution of the potassium and sodium content of the binding substance of the tuff the pH value of the solution gradually increased. For dickite to form either a higher temperature, or greater pressure or some other specific condition is needed. The fact that this mineral is present in a subordinated amount shows that all these circumstances were lacking. Usually nacrite forms at a lower temperature this is also supported by the fact that it often occurs together with cinnabar. In the southern part of the Tokaj-Mountains Hg does not even appear as trace element. Apparently for the formation of nacrite too quite specific conditions are required.

Regarding the appearance of alunite the question of the role of

potassium arises. The changing of the acidity is favourable for the weathering of the feldspars from where potassium may be dissolved, but in such cases it is also favourable if the glassy component in the rock contains potassium. Potassium may also be dissolved from deeper layers. In the deposits this must all be taken into account. It is, however, a fact that the amount of K may have been quite insignificant which is demonstrated by the local appearance of alunite and the complete lacking of illite.

The appearance of the calcite shows that the pH value of the solutions rises slowly and has shifted towards the alkaline region. However, it did not increase to such an extent for the physico-chemical conditions to be suitable for the formation of zeolite.

The lacking of the illites proves that the potassium needed for their formation was already removed when the environment would have become suitable for their formation.

The passages of the breaking out hot water solutions are bound to fault lines thus of course, the most intensive kaolinization will be found along these lines. The kaolin bodies with irregular shapes will form along them. The formation of the bentonite deposits is not unfailingly bound to the fault lines their greater part forms from tuffite of rhyolite, respectively.

Bodrogszegi

In rhyolite tuff of Sarmatian age stocklike kaolin occurrences with about a 200 m long trend and 30—50 m wide at about a depth of 25—50 m are known as has been recently described by *Frits* [1956]. The geological position of the formation is not yet quite elucidated. The underlaying rock is formed of pumicestone rhyolite tuffs with a »stone powder« texture. Perlite and obsidian lapillies varying in colour occur in it. It hardly contains any coloured minerals. In the vitreous binding substance splintered quartz crystal fragments may be detected. Generally the tuff is finely granular and the deposite is not divided.

The substance of the kaolin body is fairly uniform and the finely granular substance is permeated by limonitic manganic veins (Fig. 10.).

The rhyolite tuff is boundered by quite irregular surface. In some places swelling outs can be observed. Both the kaolin and the bordering tuff part is irregularly and networklike permeated by a light brown substance which is merely contaminated kaolinite stained with limonite and fireclay, respectively. This can also be proved by the fact that if this substance is pulverised, the iron may easily be dissolved with diluted HCl and a yellow white substance remains, the DTA curves of which show the curves characterizing these substances. Between the kaolin body and the tuff, in some instances in the kaolin body too, there are sliding planes which are sometimes brown coloured. Under thermicroscope in the matrix of the tuff appendixlike formations could be found which proved to be kaolinites. They disintegrate easily into smaller fragments vertically to their longitudinal axis, hence they can only form locally because they do not bear any transport.

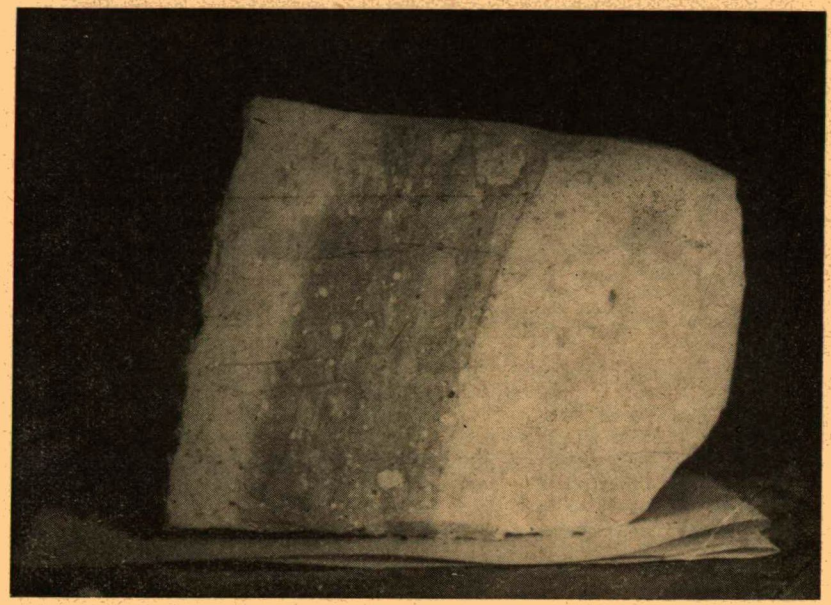

Fig. 10. Kaolin stained with limonite bands.

The grain-size analysis of the substance of the kaolinbody furnished the following result:

Table 3.

	0,25 mm ⊘	0,25—0,5	0,5—1,0	1,0
V. level, SE part	87,50 %	7,31 %	0,48 %	4,71 %
V. level, SW part	73,80	24,16	1,74	—
V. level, S part	94,20	3,10	2,70	

In detail the grain-size analysis shows that the size of those not exceeding 0,02 mm amounts to about one quarter of the substance, hence the mechanical analysis also supports the fact the substance contains very many colloidal fractions and that the single samples do not contain fractions exceeding one mm in diameter.

Just in the case of the minerals belonging to the kaolin group the DTA examinations do not give an unequivocal result. The examinations show that the substance surely contains kaolinite this is also proved by photomicrographs and electronmicrographs. The differentiation of halloysite and fireclay by means of DTA is doubtful owing to the nearly identical dehydration sections. In the curves the endotherm peaks between 100—200° C developing a different manner, generally they appear between 130—140° C which is according to the data of the literature characteristic of fireclay. The DTA examinations of the country rock only show an endotherm peak at about 570° C which characterizes kaolinite, whereas the exotherm peak is only weak (Fig. 11.).

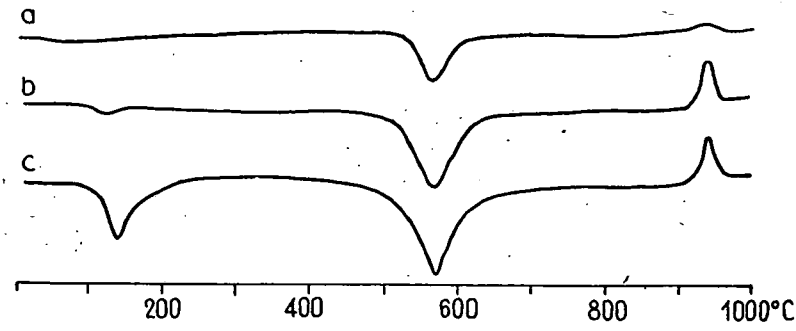

Fig. 11. a) The DTA curve of the country rock of the kaolin body.
b) The DTA curve of the substance of level IV.
c) The DTA curve of the substance of level V.

To identificate the substance exactly, Debye-Scherrer diagrams were prepared from the substances of various levels and fractions.

Table 4.

1		2		3		4		5		6	7	8
d (kx)	I	d	I	d	I	d	I	d	I	d	d	d
7,3	vw	7,33	9			7,24	10	7,3	7			
4,9	vw			7,04	8					7,15	7,17	7,17
4,46	w									4,455	4,455	4,47
		4,315	10	4,370	10	4,370	10			4,35	4,36	
								4,16	10			
4,18	vw									4,17		
3,59	w	3,568	8	3,601	8	3,592	6	3,602	7	3,566	3,57	3,577
3,38	vw									3,365		
		3,277	4	3,284	6							
								3,191	4			
								2,995	2			
2,76	vw									2,748		
2,64	w											
		2,511	8	2,528	10	2,540	6	2,515	6	2,521	2,50	
		2,457	5									
2,35	w	2,319	8	2,320	10	2,324	6	2,315	6	2,331	2,325	2,336
								2,175	1	2,182		
2,01	vw											
		1,973	3	1,9855	5	1,9805	1	1,9473	3	1,985	1,977	1,985
				1,8000	4			1,8312	2	1,835		
		1,783	1							1,778	1,785	1,786
1,693	w									1,682		
		1,660	6	1,6615	8	1,658	5	1,6692	4	1,659	1,659	
		1,6126	2	1,608	2					1,616		
		1,5304	2	1,531	3					1,539	1,541	1,538
1,496	w									1,486	1,486	1,485
		1,4785	8	1,4720	10	1,4735	7	1,4754	6	1,464		1,455
		1,4445	2	1,4395	2					1,449		
1,293	w	1,3656	3	1,3656	4							1,372
		1,2755	4	1,2765	6	1,2758	4	1,2802	3			1,2847
1,242	w											1,2476
		1,2288	4	1,2306	6	1,2242	3	1,2270	3			

The data of the exposure of the X-ray diagram:

1. Fe Kα without filter, 30 kV 12 mA.
2, 3, 4. Co anticathode, Fe filter, 23 kV, 32 mA.
5. Cu anticathode, Ni filter, 27 kV, 4,2 mA.

The samples were taken from the following places:

1. Bodrogszegi, V. level, original powdered substance,
2. Bodrogszegi, V. level, original powdered substance,
3. Bodrogszegi, V. level, a fraction of 0,002 mm of the previous substance,
4. Bodrogszegi, V. level, a fraction not exceeding 0,002 mm,
5. Bodrogszegi, IV. level, a fraction not exceeding 0,002 mm,
6. Kaolinite values obtained by *Brindley* and *Robinson,*
7. Fireclay values obtained by *Brindley* and *Robinson,*
8. Fireclay values obtained by *Robertson, Brindley* and *Mackenzie.*

On comparing the values concerning kaolin and fireclay reported by *Brindley* and *Robinson* [1951] and by *Robertson, Brindley* and *Mackenzie* [1954] with our data relating to fireclay, in both cases the values are in fairly good agreement. The fact that the kaolinite values relate to the well crystallized kaolinite explains why in this case relatively fewer lines appear. In addition some lines refer to the presence of feldspar and quartz. The lacking of well crystallized kaolinite is shown by the absence of the very characteristic lines 4,17 and 4,12. The 2,32 *d*-value referring to the fireclay appears in most of the samples very intensively.

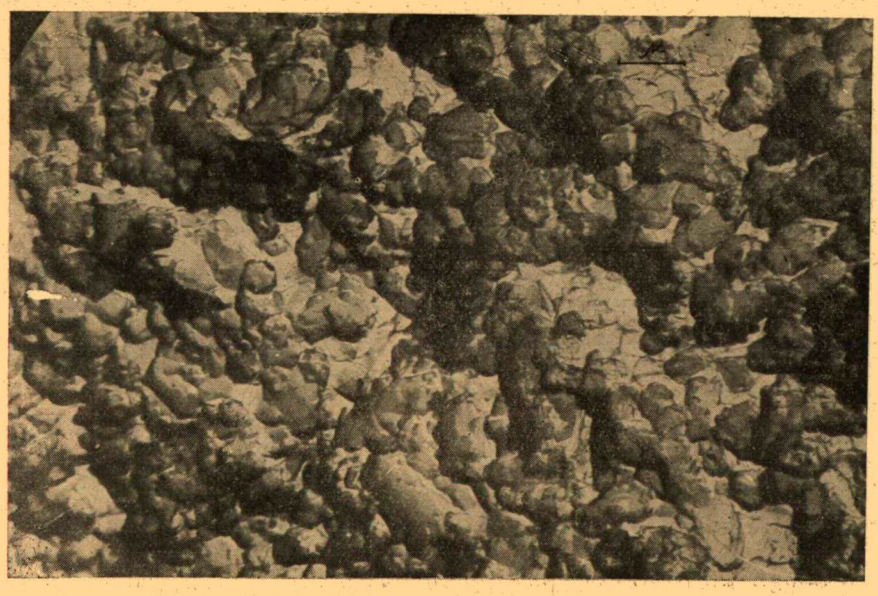

Fig. 12. Electron micrograph of the substance from level V prepared with the replica method.

The evaluation of the X-ray data also only shows that beside kaolinite fireclay is certainly present. However, the colloidal character is demonstrated by the fact that the lines are fairly wide and in some cases blurred. It is also striking that the very intense lines are nearly all lacking. Hence the substance is only composed of weakly crystallized kaolinite and fireclay.

The electron micrographs, at least those relating to the substance of level V, exclude halloysite. On the replica photos distorted hexagonal formations pointing to kaolinite can only rarely be seen. However, it may very well observed that the substance contains many colloidal fractions consequently globular-rheniform forms appear (Fig. 12).

Table 5. illustrates the data of the chemical composition recalculating the analytical results to elements and the minor elements.

Table 5.

	1	2	3	4	5	6
$Si^{4+} \cdot 10^2$	2276	2144	2661	2156	2160	1996
$Al^{3+} \cdot 10^2$	1670	1746	1312	1797	1778	1692
$Ti^{4+} \cdot 10^2$	11	7	6	7	6	13
$Fe^{3+} \cdot 10^2$	194	213	147	201	206	197
$Fe^{2+} \cdot 10^2$	13	6	24	10	10	—
$Mn^{2+} \cdot 10^2$	2	4	4	3	2	6
$Mg^{2+} \cdot 10^2$	10	—	—	—	2	2
$Ca^{2+} \cdot 10^2$	14	13	34	16	14	25
$Na^{1+} \cdot 10^2$	7	7	32	8	7	25
$K^{1+} \cdot 10^2$	42	32	213	31	31	26
$P^{5+} \cdot 10^2$	3	2	1	2	2	7
$S^{6+} \cdot 10^2$	—	—	—	—	—	3
B						(tr)
Cu						tr
Pb						tr
Si/Al	1,36	1,23	2,03	1,20	1,21	1,19
Al/Fe^{3+}	8,61	8,19	8,93	8,94	8,63	8,48
$Na + K \cdot 1000/Al$	29,34	22,34	186,74	21,70	21,37	30,50
K/Na	3,00	4,57	6,66	3,87	4,43	1,04

The occurrences of the samples:

1. Boring II. 28,3—31,7 m,
2. Boring II. 43,9—47,3 m,
3. Boring II. 63,0—66,4 m,
4. Boring II. 72,6—75,3 m,
5. Boring II. 81,9—84,1 m,
6. Paperkaolin.

Although the coloured rock-making minerals do not play an essential part in the rock, yet the relatively high value of Fe^{3+} is striking. This high iron content is also demonstrated by the fact that limonite

often stains the kaolin in stripes this can be particularly well observed on the V. level of the South-Western area. In this part manganese minerals are not present they may rather be found on the part in the vicinity of the tuff. In the places where manganese minerals occur the kaolin is nearly quite pure, the manganese compounds are concentrated in small groups, whereas the compounds containing iron mostly stain the kaolin yellow.

The chemical composition of the substance is fairly uniform. This holds good too for the parts which are also worked at present and for the substance of the borings. According to the analytical data the Si/Al ratio is generally higher than ideally in the case of the kaolin.

The $K + Na \cdot 1000/Al$ value shows the amount of the feldspar which remained intact. At the measuring of the grain-size it was revealed that the greater part of the substance from Bodrogszegi can be suspended. In the suspension most of the grains belong to the clay fraction. If it is taken into account that this factor diminishes with grain-size its low value may be understood. But for one exception the values are approximately identical which means that the kaolinization is about uniform in the whole deposit.

Under certain conditions the potassium sodium ratio indicates the relation between two different feldspars. In view of the fact that the kaolin minerals have a low adsorption capacity the potassium and sodium can only originate from the not yet weathered feldspar and to a smaller extent from the glassy pumice-stone binding substance. The ratio generally exceeds one, showing that the sanidine content of the altered rock is also in the present state larger than the amount of the plagioclases, i. e. the potassium content of the binding substance is higher than that of the sodium.

According to *Robertson, Brindley* and *Mackenzie* [1954] who have dealt in detail with investigation of kaolinite and fireclay chemical calculations have shown that in the fireclay type in the tetrahedral layers the substitution of the $Si \rightarrow Al$ is the double of that in kaolinite and the members of the kaolin groups with a regular structure in general. While on the one hand, in the tetrahedral layers such differences can be distinguished in the various types, on the other hand, in the octahedral layer such differences cannot be found in the substitution. In our case this could not be established because pure substance was not available thus the amount of colloidal silicic acid is not known and a decision wether or not there is Al in excess could be reached.

The minor element-associations show a very scanty picture. Of the chalcophil elements only Cu and Pb appear in traces and of the sedimentophil ones only B is present with a very weak spectral line. Whilst the amount of Cu and Pb ranges between 0,01—0,001 per cent, the amount of B does not exceed 0,00001 per cent, it is, therefore, quite insignificant.

The occurrence of fireclay in the Tokaj-Mountains is not extensive in kaolins it can only be found here. On the basis of the data of

the literature fireclay is a mixture of the refractory clay minerals. So it appears in Pilisszentiván where the parent-rock of the refractory clay containing fireclay is probably an lateritic sediment. In the refractory clay from Cserszegtomaj also occurs the kaolin of the fireclay type. In this case the kaolin and bauxite formations are probably due to the same substance. Only the different physico-chemical conditions altered the environment and rendered possible the associated occurrence of the two formations.

Robertson, Brindley and *Mackenzie* [1954] established that the crystals from Pugu irregular arranged in the direction of the *b* axis are morphologically very perfect. They believe that the cation exchange capacity is rather due to the tetrahedral and octahedral positions than to the valencies of the crystal edges. In the deposit of Pugu migmatite and gneiss are the original rocks. The irregular kaolin crystale arranged according to axis *b* represent together with a finely granular quartz and feldspar sand a deeper sea facies.

In Bodrogszegi the formation conditions were different. The appearance of the andezite volcanity rendered the keeping open of the lines possible to this the formation of a cleavage structure following the orogenic movements contributed too. According to the traces along the fault lines very weak hydrothermal solutions broke out at a very low temperature. This process could, however, not be permanent, which is also proved by the fact that although the feldspars decompose, the residue is usually composed of colloidal substances X-ray analyses and electron micrographs also demonstrate this. The kaolinite present also demonstrates that epigen effects play an essential role at the formation. The appendixlike kaolinites are very weakly crystallized. The appearance of colloidal substance in larger amounts suggests that the hydrothermal solutions only play a greater role at the decomposition of the feldspars. The weak crystallinization of the kaolinites furthermore, the presence of fireclay prove that epigen effects played a great role in the formation of the deposit. Te pH value of the circulating solutions was throughout low as minerals, on the basis of presence of which a greater fluctuation of the pH might be assumed, could not be found.

Concerning the formation of the fireclay all data emphasise the great role played by the epigen conditions. In this case, therefore, according to the above this must primarily be taken into account, i. e. the hydrothermal solutions only performed the breakdown of the feldspar and the transport of the released ions. The formation of the new minerals is mostly due to epigen effects.

The continental origin of the transformed tuff is also proved by the absence of the montmorillonites which are represented in the vicinity in fairly large amounts and furthermore, that no layers can be detected in the tuff. The absence of illites points to the fact that the pH value of the solutions did not change appreciably during the transformation furthermore, that during this period the circulation of the solutions was also good and the K ions were transported as a whole.

Szádeczky-Kardoss refers to the fact that in the magmatic processes the liquid magmatic section composes as compared to the resudual magmatic phase a separate system. The separation sequence of the rock-making minerals also shows decreasing compound potentials. In the residual solutions on the basis of the decreasing compound potentials. It the residual solutions on the basis of the decreasing compound potentials a sequence may also be established which also corresponds on the whole to the formation sequence. Hence, both at the crystallization of the solutions as well as at the mineral associations formed on the action of the hydrothermal solutions decreasing compound potentials are obtained. After the liquid magmatic phase suddenly another series showing a decreasing compound potential starts. At the weathering the experimental sequence of the secondarily formed clay minerals can also be ranged according to the decreasing compound potentials.

Using the ion and anion potential values of *Szádeczky-Kardoss—Grasselly* [1959] the minerals forming at the alteration due to the hydrothermal solutions may be ranged according to the decrease of their compound potential:

Table 6.

minerals	composition	compound potential
Or-adular	$KAlSi_3O_8$	0,72
quartz	SiO_2	0,71
kaolinite	$Si_3AlO_{10}Al_4(OH)_8$	2,07
	$Si_4O_{10}Al_4(OH)_8$	2,06
alunite	$KAl_3(OH)_6(SO_4)_2$	2,02
illite	$KAl_2Si_4O_{10}(OH)_2$	1,94
montmorillonite	$Si_8O_{20}(OH)_4Al_3Mg$	1,91
beidellite	$Si_8O_{20}(OH)_4AlFe_3$	1,87
nontronite	$Si_8O_{20}(OH)_4Fe_4$	1,79
calcite	$CaCO_3$	1,42
baryte	$BaSO_4$	1,03

Of course, these compound potential values were calculated from a simplified formula not taking into account the substitution possibilities although there are wide possibilities for the substitution in the case of the phyllosilicates on the one hand in the tetrahedral layers, which in most cases means a Si — Al substitution, and on the other in the octahedral layers between Al—Mg, Al—Fe^{3+} etc. The above compound potential values show clearly that the members of the montmorillonite group, apart from the extreme values, succeed illite and kaolinite.

The values of the above table can be plotted in a coordinate system where on the one axis the approximate pH intervals and on the other the ψ values are shown. The succession is well visible.

At the formation of these minerals the change of pressure is not essential as the process occurs on the surface or near it. The change of temperature must already be taken into account, however, this also ranges between relatively small limits, usually it decreases gradually. Concluding from the minerals formed and from the present temperature of the solutions there has not occured any considerable change in

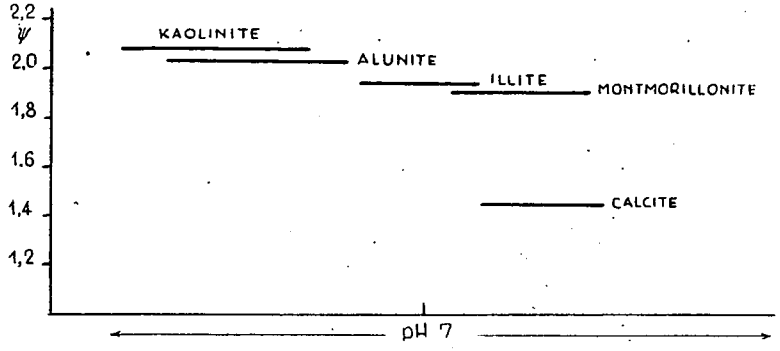

Fig. 13. The mineral association of Végardó as a function of ψ and pH.

their temperature in the Tokaj-Mountains it ranges between 200—230° C. Only the change in the concentration of the solutions was significant. The pH value increased gradually from 2—3 to about 8—9. Thus, at the development of the types of the different minerals the change in the concentration and in the pH played the most important role. The decreasing compound potential values quasi indicate what possiblities existed under the given conditions at the formation of the different minerals. The rare occurrences of beidellite and nontronite may be explained by the fact that in some cases it was not possible that besides the other conditions of the formation iron should also be present in considerable amounst.

The succession of the minerals separating from the solution can be characterized by the Bowen series. In this case the compound potential values show a decreasing value. The orthoclase and quartz, however, separating at the end show an appreciably lower value than the kaolinite or alunite forming first on the action of hydrothermal solutions. The series of the minerals forming after kaolinite is a series according to a decreasing compound potential into which apart from the clay minerals both calcite and baryte, but even chalcedon, fit.

Szádeczky-Kardoss demonstrates in the case of the magmatic processes two separate systems, the compound potential values of which also differ distinctly from each other. The one is the liquid magmatic phase and the other the residual-magmatic phase. The clay mineral series forming secondarily on the action of the hydrothermal solutions again shows a sudden change in its compound potential thus from that point of view this is a third phase. Like in the postmagmatic phase in this case also a great amount of solvent is present and the concentration of the ions is very low.

Taking the Si/Al ratio of the decreasing values of the above compound potentials into account, beginning from the kaolinite towards illite and montmorillonite the ratio gradually shifts and the amount of the Si gradually increases. In this series too like in the Bowen series the decreasing compound potential value indicates an increasing degree of silification. This is well illustrated in Fig. 14.

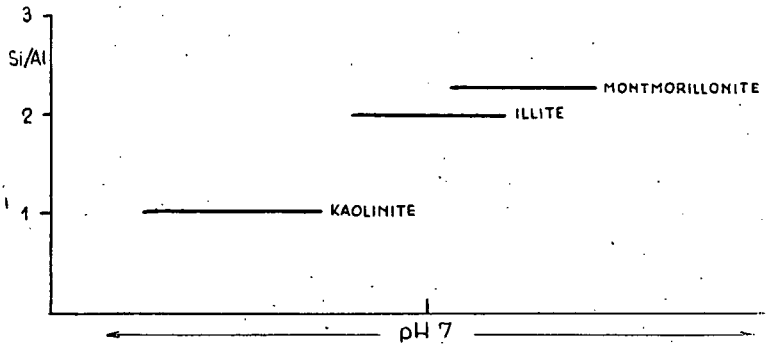

Fig. 14. The Si/Al ratio of clay minerals as the function of the pH.

In the examined cases the kaolins of the Tokaj-Mountains are poor in minor elements. The kaolinites from Mád, Bodrogszeg, Sárospatak and Végardó characteristically only contain Cu and Pb. The alunite occurrence of Végardó is an exception as in it V, Cr, Co, Ni, Sn, Mn, and As also appear in traces. Pyrite also only contains few minor elements. The purity of the spectrum demonstrates that both the pyrite appearing crystallized and the pyrite in opal is an epigen formation. The compound potential value fits well into the series of the clay minerals. As according to *Szádeczky-Kardoss* two compounds can only crystallize together if their compound potential is similar, the fact that pyrite also forms after the clay minerals may be explained on the basis that its compound potential is far lower than the one of kaolin, besides other circumstances this also points to the fact that these two minerals cannot be syngenetic.

The theory concerning the above sequence of formation of the clay minerals is also in agreement with the examinations of Korshinsky. According to him the acid components of a solution (CO_2, HCl, H_2S etc.) penetrate far quicker into a rock than the other components of the solution. In aqueous solutions the acid components have an ion mobility which is greater on the average than that of the alkaline components. Particulary the H ion is far more mobile than the OH one. Hence, it may be assumed that the acid components the ascendent solutions penetrate far more quickly upwards than the alkaline components. Consequently the acidity maximum of the postmagmatic solutions must be less deep, in abyssic depth this condition shift towards the alkaline region. This is the reason why in greater depths sericite, and in higher regions kaolin, take the place of feldspar.

This is also expressed by the establishment of *Szádeczky-Kardoss* concerning the volatiles. According to him in not great depths only the most light volatiles, water, CO_2 and sulphur originating from organic substances exert a transvaporisation action.

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THE CHANGE OF THE ELECTRODE POTENTIAL OF SPHALERITE-PYRRHOTITE SYSTEMS AND ITS ROLE AT THE WEATHERING OF SULPHIDE ORE DEPOSITS

(Preliminary Report)

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INTRODUCTION

The outcrop of sulphide ore deposits and their part beneath the surface, respectively, is exposed to the effect of chemical and physical factors of the weathering. The study of the oxidation processes taking place in the oxidation zone is essential also from the point of view that the cementation zone, often representing economically important and valuable mineral enrichment, forms in the deeper part of the ore body due to the action of metalbearing aqueous solutions seeping down from the oxidation zone.

The processes taking place at the weathering of sulphidic ore deposits earlier were interpreted as simple chemical reactions, the electrochemical properties of the single ores, however, were only less taken into consideration. The relatively few referring data of the earlier literature are comprised by *H. Schneiderhöhn* [1924] in his comprehensive work dealing with the mineralogy of oxidation and cementation zone, respectively.

The examinations mentioned above have dealt partly with the solubility relations of the single ores and the interaction referred to the solubility due to the contact of the ores, partly with the problem of the electromotive force series of the sulphide ores.

It is plausible that the solubility relations of the single ores are essential at the weathering of the ores and the formation of the oxidation and cementation zone. In the change of the solubility properties, however, the differences among the electrode potentials of the single ore minerals play an important role.

Recently, examinations regarding to the electromotive force series of these ores are carried out by *Rechenberg* [1951]. He used as electrolyte, not destilled water as *Gottschalk* and *Buehler* [1910], but solutions of different concentration and pH. On the basis of his examinations he stated that apart from minimal deviations the sulphide ore minerals in a given e. m. f. series can be arranged and the sequence of the minerals in this series is determined by structural properties. *Rechenberg*'s opinion: the sequence according to the potential, recalculated to the normal hydrogen electrode, is analogous with the normal potential series of the metals.

The question is, whether it would be possible to establish a defined potential series of general validity referring to the sulphide ore minerals or not, and further, how far would such a series be applicable to interpret the weathering relations of single sulphidic ore deposits.

It is pointed to by *Grasselly* [1953] that while the potential series of the metals means the series according to the normal potentials, measured at exactly defined conditions, the normal potential of natural sulphide ore minerals can not be so simply established. The sulphide ores approach only exceptionally the ideal composition containing nearly always mechanical and/or isomorphous impurities in varying quantities, further, they are mostly in contact not alone with self-ionic aqueous solutions and the ion-concentration is also changeable. It seems to be more reliable studying the weathering of sulphide ore deposits that not so much the establishing of a defined potential series of general validity for sulphide ore minerals is essential, as the establishing of the potential series valid for the given deposit under the given circumstances, putting up this series by taking into account of the actual circumstances of the given deposit such as its mineral association, the actual composition of the ore minerals, their mechanical inclusions and isomorphous impurities, the pH and the composition and concentration of the aqueous solutions being in contact with the ores.

According to the above-mentioned, the potential of the single ore minerals gives some explanation regarding their solubility relations only in the case if these potential values are not taken into consideration in general, but are referred to the environment wherein the ores occur, because also the same ore mineral in an other occurrence can show partly an other potential value, partly also at the same potential value a different behaviour can be observed owing to the different environmental conditions.

Recently, *Noddack, Wrabetz* and *Herbst* [1955a, 1955b, 1956] have treated the electrode potential of natural and artificial sulphides not so much from geochemical but mostly from physico-chemical viewpoint.

According to their examinations most of the natural and artificial sulphides follow the *Nernst's* equation and in a galvanic cell, building with their own metal, they represent the positive pole. On the basis of the measured potentials, a defined electromotive force series is given

also by these authors. It is to be noted that in the series both natural and artificial sulphides are present. The authors pointed to that the results are strongly influenced by stoichiometrical deviations and by the presence of oxygen either on the surface of the electrodes or in the electrolyte.

The effects influencing the electrode potentials of the sulphide ore minerals are derived partly from the properties of the ore minerals themselves, partly from the properties of the environment. Thus, from the point of view of the electrochemical processes playing an essential role at the weathering, among others, also the changes in the composition of the ores, being caused either by mechanical inclusions or by isomorphus components, are to be taken into account.

THE EFFECT OF THE CHANGE OF THE Zn/Fe RATIO ON THE CHANGE OF THE ELECTRODE POTENTIAL OF SPHALERITES

The sphalerite-pyrrhotite system seemed to be very suitable to study the potential-influencing role of the isomorphous components, since partly the iron-containing sphalerite is common ore mineral of hydrothermal sulphide deposits, partly as in the sphalerite the zinc can be substituted by iron in considerable amounts and this solid solution in given cases may remain stable also by normal temperature. The iron content of the sphalerites depends upon the pressure and temperature at the time of its formation. The higher the formation temperature the greater amount of the zinc can be substituted by iron in the lattice of the sphalerite.

The question is whether the pyrrhotite content and its change in the sphalerites causes changes in the electrode potential of sphalerite, and if it does, in what direction this change takes place. The purpose of the examinations was to establish the connection between the change of the pyrrhotite content and the electrode potential of the sphalerites, the direction of the probable change of the electrode potential and further to establish whether this change is unequivocal or not with the change of the iron content.

The measurements were carried out by Orion pH electrometer. The electrodes were sphalerite specimens from various localities. The Zn and Fe content of the samples were determined. Electrolyte was $ZnSO_4$ solution in various concentration. Reference electrode: n calomel electrode. The potential values in the Tables are those measured *vs.* n calomel electrode. At the grinding and polishing of the surface of the ore sections care was taken to have possibly identical surface quality in the case of each sample. The polished ore sections were also microscopically examined to find the part on the surface where it contains no or relatively few inclusions and thus to find the part designed to take up the cell containing the electrolyte.

The locality, the zinc and iron content and their ratio, respectively, as well as the measured potentials (*vs.* N. C. E.) of the samples are comprised in Table 1.

Table 1.

Locality	Zn %	Fe %	Zn/Fe	Potential in mV 0,01 m $ZnSO_4$ pH 2,5
Rodna (Óradna, Rumania)	48	14,4	3,3	— 138
Herja (Kisbánya, Rumania)	42,8	12	3,5	— 78
Rodna (Óradna, Rumania)	54	11,9	4,5	— 105
Gyergyóholló (Rumania)	60,3	4,2	14,4	— 60
Gyöngyösoroszi (Hungary)	63	2	31	+ 230
Dognacea (Dognácska, Rumania)	52,4	1,7	31	+ 485
Baňská Štiavnica (Selmecbánya, Czechoslovakia)	63	1,8	35	+ 380
Picos del Europa (Spain)	66	0,33	200	+ 620

The data — despite of the relatively few samples — offer the possibility to answer the question. It can be stated that in the potential values of the sphalerites significant differencies prevail presumably depending upon the iron content. The potential is more positive at low iron content whereas at higher iron content the potential is less positive or even negative *vs.* the normal calomel electrode. The potential becomes more positive with increasing Zn/Fe ratio.

However, at the same time can not be exactly stated that the change of the potential would be determined only and in every case, predominantly by the change of the iron content. There were samples, the potential of which, despite of the similar iron and zinc content and Zn/Fe ratios, respectively, were different. Slight or more considerable individual differencies in potentials of the samples with similar iron and zinc content, however, do not change the tendency that the sphalerites with higher iron content, referring to higher formation temperature, posses in general rather negative potential whereas the sphalerites formed at lower temperature containing less iron, show an increasing positive potential with the decreasing iron content *versus* the normal calomel electrode.

The change of the potential of the sphalerite samples is in connection besides the iron content with other factors too, as e. g. the differencies in granular structure of the specimens, the quality and quantity of the foreign inclusions at the surface of the polished ore section. The potential of the various sphalerite samples, is co-controlled by these factors too. It is also to be taken into consideration that the iron content of the sphalerites can be present not only as FeS in solid solution, but also as unmixed pyrrhotite lamellae or drops in the sphalerite. In the latter case the presence of several micro local elements at the surface of the polished ore section is to be taken into account and also these exert an influence on the average potential of the sample. That is, two sphalerite samples with the same iron content can posses different potential depending upon whether the iron, as solid solution or unmixed pyrrhotite inclusions, is present.

The electrode potential of sphalerites of different iron content changes between — 138 mV and + 620 mV *versus* the normal calomel electrode.

THE POSITION OF SPHALERITES FROM DIFFERENT LOCALITIES IN THE POTENTIAL SERIES OF SULPHIDE ORE MINERALS

It was already mentioned that several authors have determined a defined potential series for the sulphide ore minerals. From some point of view, however, may be questionable the suitability of putting up of such a potential series of natural sulphide ore minerals, considering this series as generally valid, and further, the rightness of the interpretation of the weathering of different sulphide deposits on this basis.

What does the position of single ore minerals in the potential series mean practically from the point of view of the weathering? It means that in the case of a polymineralic ore vein, the ore minerals of higher potential, in contact with ore minerals of lower potential, are rather protected from dissolution, whereas the ore minerals of lower potential behave anodic, i. e., they will dissolve and their solubility is greater than that of which could be established in the case of pure ore minerals in no contact with others.

Arranging the sphalerites from different localities on the basis of their electrode potential into the potential series proposed either by *Rechenberg* [1951] or *Wrabetz, Noddack* and *Herbst* [1955b], it can be stated that sphalerite samples can be ranged into both series in different positions, and the electrode potential and thereof the position in the series, further the behaviour of the different sphalerite specimens in contact with the other members of these series, roughly, is determined — with the reservations mentioned above — by the iron (pyrrhotite) content depending upon the locality, the conditions of the formation.

In our opinion the comparison with the potential series according to *Noddack, Wrabetz* and *Herbst* [1955b] seems to be rather reliable, though in this series besides the natural sulphides also artificial sulphides are involved, but the potentials are in self-ionic solutions measured, whereas the potential series according to *Rechenberg* [1951] contains alone natural sulphides, but the potentials are measured in solutions of foreign ions.

The fact that the various sphalerite specimens examined, in different positions can be ranged into the potential series considered of general character by the authors mentioned, may mean the following.

Table 2.

The position of sphalerite specimens from different localities in the potential series of sulphide ore minerals according to *Rechenberg* and *Noddack, Wrabetz* and *Herbst*

Potential series according to Noddack, Wrabetz and Herbst	Sphalerites, locality potential, iron content	Potential series according to Rechenberg	
	Picos del Europa 620 mV; 0,33 %		
Ag_2S (art.) 579 mV			
	Dognacea 485 mV; 1,7 %		
	Baňská Štiavnica 380 mV; 1,8 %		
		Pyrite	377 mV
		Marcasite	351 mV
CuS (art.) 319 mv			
		Chalcopyrite	277 mV
	Gyöngyösoroszi 230 mV; 2 %		
ZnS (nat.) 219 mV			
Cu_2S (art.) 199 mV			
		Sphalerite	178 mV
FeS_2 (nat.) 139 mV			
		Pyrrhotite	117 mV
		Galena	114 mV
PbS (nat.) 89 mV			
		Argentite	— 5 mV
	Gyergyóholló — 60 mV; 4,22 %		
	Herja — 78 mV; 12 %		
MnS (art.) — 121 mV			
	Rodna — 138 mV; 14,4 %		

The potential values are referred to normal calomel electrode.

While in the case of ideal or approximately ideal pure sulphide electrodes, among others in the case of ZnS electrode, a well defined potential value, and on the basis of such potential values, a defined potential series can be established, in the case of natural sulphide ore minerals as here, that is, in that of natural sphalerites of different localities and of different iron content, can not be generally spoken about *the* electrode potential of *the* sphalerite, because this potential value due to the action of several factors may be changed within wide limits. This consideration can not be limited only to the sphalerites, but it can be applied also to the natural sulphide ore minerals in general. In the nature the minerals, whose composition is close to the ideal ones are rather rare and the contamination of the ores by different foreign components is common. These mechanical and/or isomorphous impurities also in the case of the same mineral may be different depending upon the locality, the conditions of the formation and therefore, the same mineral occurring, however, in different localities,

having different electrochemical properties, from the point of view of weathering a diverse behaviour may show.

Thus considering the series of the sulphides, according to *Noddack, Wrabetz* and *Herbst* as well as *Rechenberg* in Table 2, if the potential series is considered as the potential series of the sulphides and the position of each mineral therein and the conclusions to be drawn are treated only in general, it can be stated that the sphalerite in both series is ranged approximately into the middle and the galena has a less positive potential than the sphalerite in both series, that is, in contact with galena the sphalerite will be protected from dissolution, whereas the solubility of galena becomes more considerable than if it were in no contract with sphalerite.

Inspite of all, however, can not be supposed that in contact of the two minerals mentioned above, always the galena tends to dissolved and the sphalerite will be protected as the adverse case is possible too. Considering namely, the series of the sphalerites from different localities and assuming that the composition and thereof also the potential of galena varies between closer limites than that of the sphalerite, it can be assumed that even the sphalerite with higher iron content having thus a less positive potential than the galena, will dissolve in contact with the latter. So, it is presumable that sphalerites with higher iron content, e. g. sphalerites from Rodna or Herja, in contact with galena or pyrite, being of anodic character in this mineral-association, will rather dissolve. There again, the sphalerites with low iron content, e. g. sphalerites from Picos del Europa or Dognacea, in a mineral-association of sphalerite-pyrite-chalcopyrite-pyrrhotite-galena will be protected and rather the other members of this series will be dissolved.

Thus, on interpreting the weathering processes of given sulphide deposits on the basis of the electrochemical properties of the single sulphide ore minerals, not the electrochemical properties of the single sulphides in general, but always the actual conditions and circumstances of the given occurrence are to be taken into account.

The further examinations are in progress.

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KRISTALLOGRAPHISCHE BEOBACHTUNGEN

von L. TOKODY,
Min.-petr. Abt. d. Ungarischen Nationalmuseums, Budapest.

Die weiter unten folgenden Zeilen berichten neue kristallographische Untersuchungsergebnisse bezüglich einiger Mineralien der Karpathenbecken.

MIT KRISTALLFLÄCHEN ENDENDER STALAGMIT AUS DER BÉKEBARLANG (FRIEDENSHÖHLE)

L. Vértes, Museologe des Historischen Museums im Ungarischen Nationalmuseum, schenkte im Jahre 1955 der Mineralogisch-petrographischen Abteilung des Naturwissenschaftlichen Museums einen merkwürdigen, mit Kristallflächen endenden Stalagmit aus der im Gebiet von Jósvafő liegenden Békebarlang (Friedenshöhle) des Gömörer Karstes. Dieses Stück ging 1956 zugrunde. Zum Ersatz sammelte *L. Vértes* 1959 vier ähnliche Stücke. Für seine freundliche Bemühung spreche ich ihm auch hier meinen besten Dank aus.

Die vier Stalagmite beanspruchen wegen des Auftretens von Kristallformen näheres Interesse.

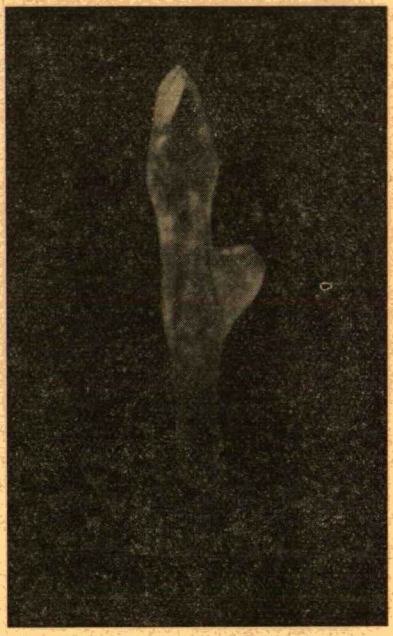
Alle vier Tropfsteine stammen von der Wand der »Kötélhágcsószifon« (»Strickleitersiphon«) benannten Partie der Békebarlang (Friedenshöhle). Die Stücke können eigentlich weder als Stalagtite noch als Stalagmite bezeichnet werden, da sie an die Wand der Höhle angewachsen sind. In ihrem Inneren befindet sich kein Hohlkanal; sein Fehlen deutet auf den Stalagmitcharakter der Tropfsteine hin.

Alle vier Tropfsteine sind milchweiss, durchscheinend. Ihr Stoff ist Kalzit. Es befinden sich unter ihnen zwei kleinere, 20 mm lange und 8, bzw. 9 mm dicke Stalagmite; am Ende des einen von ihnen sind Spuren gekrümmter Kristallflächen zu sehen.

Zwei grössere Tropfsteine verdienen ausführlicher besprochen zu werden.

Das eine Stück ist 52 mm lang; sein dünnerer, an der Höhlenwand haftender Teil ist 4 mm, das freie Ende 11 mm dick. Der freie,

dickere Teil befindet sich an einem schwach gebogenen »Stiel« und gleicht einem herabhängenden Tropfen. Am unteren Teil des Tropfens zeigen sich Spuren von gekrümmten Kristallflächen. Am Ende des Stalagmits aber, in der Richtung seiner Längsachse, sind gut entwickelte Kristallflächen. An der oberen Teil dieses Tropfsteins ist parallel ein wesentlich kleinerer Stalagmit angewachsen, und an ihm haben sich ebenfalls Kristallflächen ausgebildet. Die goniometrischen Messungen ergaben, dass diese Flächen zu der beim Kalzit häufigen Form $f(02\bar{2}1)$, — 2R gehören. (Fig. 1.)



Figur 1. Mit Kristallflächen endender Stalagmit aus der Békebarlang (Friedenshöhle).

Das zweite Stück ist 46 mm lang und 4—7 mm dick. Am Ende dieses Tropfsteins erschienen ebenfalls die Flächen der Form $f(02\bar{2}1)$. Am Ende einer 9 mm langen Anwachsung, die vom Stalagmit etwa in seiner Mitte herabhängt, lassen sich gekrümmte Rhomboederflächen beobachten; sie entsprechen wahrscheinlich ebenfalls den Flächen der Form $f(02\bar{2}1)$.

Die Flächen der Form $f(02\bar{2}1)$, die am freien Ende beider Tropfsteine auftreten, sind trüb, ihre Reflexion ist schwach, sie erlauben aber die sichere Bestimmung der Form. Die gekrümmten Flächen eignen sich nicht zu Messungen.

Das Wachstum der Tropfsteine mit Kristallflächenendung ist äusserst langsam vor sich gegangen, zumindest an den Teilen, wo sich die Kristallflächen ausgebildet haben.

Über Tropfsteine mit Kristallflächenendung habe ich im mineralogischen Schrifttum keine Angabe gefunden. Somit sind die Tropfsteine der Békebarlang (Friedenshöhle) besonders beachtenswert.

Speleologisches Schrifttum stand mir bloss in ganz beschränktem Masse zu Verfügung. *Trimmel* (1) beruft sich in seiner Abhandlung auf die Arbeiten von *Anciaux* (2), *Moore* (3) und *Geze* (4); sie beschreiben unter dem Namen »Excentriques«, bzw. »Heliktites« aus Kalzit oder Aragonit bestehende dünne Stalagmite, die der Form nach als vermiformes (wurmförmig), filiformes (fadenförmig) bezeichnete Gebilde sein können. Unter ihnen kommen zuweilen auch Stalagmite vor.

Trimmel führt die Höhlen von Österreich an, in denen er »Excentriques« beobachtet hat. Aus diesen Höhlen erwähnt er aber keine Tropfsteine mit Kristallflächenendung.

Die Arbeit von *Kunský* (5) berichtet weder in den Beschreibungen noch mittels der Fotografien über Gebilde, die sich mit den Tropfsteinen der Békebarlang (Friedenshöhle) identifizieren lassen. Auf einem Lichtbild (ohne Seite- und Tafelangabe) sind in der Höhle »Na Zlatém koni« (am goldenen Pferd), die südwestlich von Prag, im Kalkstein des Böhmischen Karstes, unweit der Stadt Beruon, bei dem Dorf Koněprusy liegt, Stalagmite zu sehen, die an die Tropfsteine der Békebarlang (Friedenshöhle) erinnern, doch sind auch an ihren Enden keine Kristallflächen erkennbar.

BERYLL VON MACSKAMEZŐ

Im Gebiet der Karpathenbecken ist der Beryll ein selten vorkommendes Mineral. Zwei Beryllfundorte sind bekannt: Teregova und Macskamező (Răzoare [Mașca]).

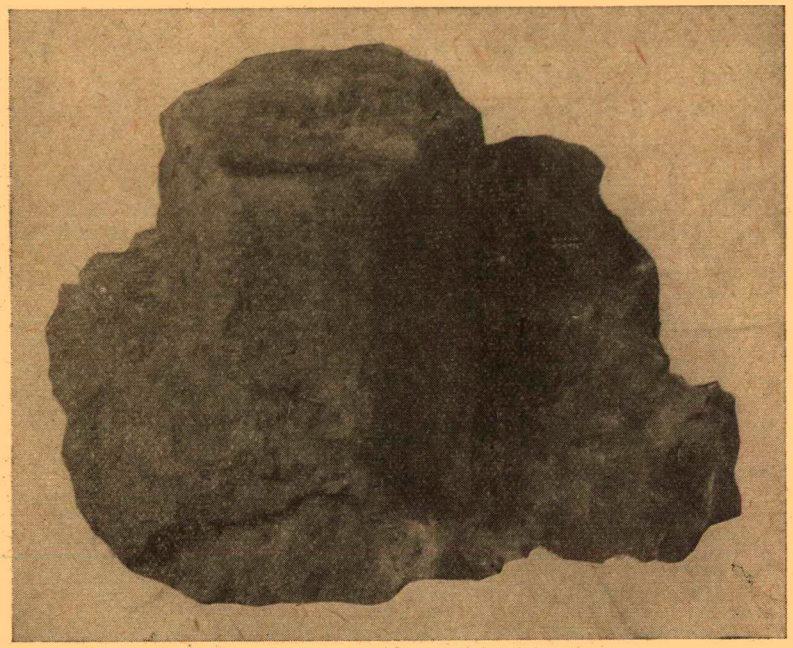
Der Beryll von Teregova ist zuerst bei *Schadler* erwähnt (6). Mit seiner ausführlichen Untersuchung befassten sich *Dittler* und *Kirnbauer* (7, 8).

Die Mineralien von Macskamező wurden von *Zsivny* beschrieben (9). Vom einzigen gefundenen Beryllstück teilte er die folgenden Daten mit. Länge des Kristalls 8,5 cm, Dicke 3,7—4 cm; festgestellte Kristallform: hexagonale Prisme I-ter Ordnung; Farbe: blassbläulichgrün; H: zwischen 7 und 8; $n = 1{,}57$; ist mit glimmerhaltigem Quarz verwachsen.

Zsivny teilte ausserdem auch seine Beobachtungen über den Turmalin, Granat, Kalifeldspat, Muskovit und Biotit mit.

L. Székely, Oberingenieur des Montankonstruktionsbüros, schenkte dem Ungarischen Nationalmuseum im Jahre 1958 mehrere Minerale von Macskamező, grösserenteils Glimmer- (Muskovit-) Platten. In mineralogischer Hinsicht verdienen aber mehr Beachtung die einzigen Exemplare je eines Beryll- und Muskovitkristalls sowie ein von Spaltungsflächen begrenzter grosser Orthoklas.

Der Beryll ist teilweise in Granitpegmatit eingewachsen. Der nach $(10\bar{1}0)$ etwas flache Kristall hat eine bedeutende Grösse; seine Höhe beträgt 74 mm, seine Breite 49 mm, seine Dicke 41 mm. Er ist auf dem Lichtbild in nahezu natürlicher Grösse dargestellt. (Fig. 2.)



Figur 2. Beryllkristall von Macskamező.

Seine Kristallformen sind: c(0001), m$(10\bar{1}0)$ und a$(11\bar{2}0)$.

Die Form c(0001) hat sich bloss am oberen Teil des Kristalls ausgebildet. Sie erscheint nicht als eine einzige Fläche, sondern bildet sich unter Wiederholungen aus. Sie ist etwas brüchig. Stellenweise ist die zu ihr parallele schlechte Spaltung erkennbar.

Die herrschende Form des Kristalls ist m$(10\bar{1}0)$. Ihre Flächen sind die besten Flächen des Kristalls. Wie oben erwähnt, beobachtete *Zsivny* am Beryll von Macskamező bloss die Form m$(10\bar{1}0)$.

Die Form a$(11\bar{2}0)$ erschien mit zwei Flächen, mit $(11\bar{2}0)$ und $(2\bar{1}\bar{1}0)$, mit der letzteren bloss in Spuren. Die Ausbildung dieser Flächen ist nicht so gut, wie die der Form m$(10\bar{1}0)$. Die Oberfläche ist rauh. Die Kanten m: a sind nicht scharf.

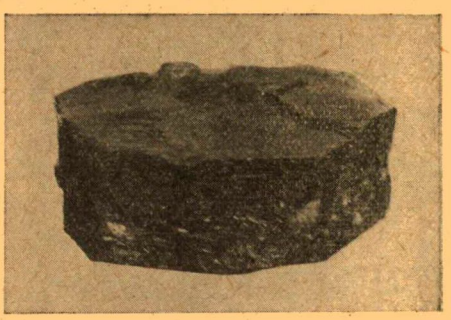
Der Habitus des Kristalls ist wegen des Vorherrschens der Zone [0001] säulig.

Die physikalischen Eigenschaften sind charakteristisch für den Beryll: erkennbare Spaltbarkeit nach (0001), muscheliger Bruch, Härte 7,5 blassblaue Farbe, Undurchsichtigkeit, Glasglanz.

MUSKOVIT VON MACSKAMEZŐ

Zsivny (9) berichtet über den Muskovit von Macskamező bloss, dass er ihn in grossen Tafeln gefunden hat und dass das Mineral häufiger vorkommt als der Biotit.

In der Schenkung von *L. Székely* befinden sich 20—25 cm grosse Muskovittafeln mit unbestimmten Konturen, Biotit nur in einem Exemplar mit tafeliger Ausbildung.



Figur 3. Muskovit von Macskamező.

Beachtenswert ist ein Muskovitkristall. Die Fotografie (Fig. 3.) des etwa 50 mm breiten und 15 mm dicken Kristalls ist auf der Abbildung zu sehen. Seine Kristallformen sind: c(001), b(010) und m(110); von diesen begrenzen (001), (0$\bar{1}$0), (110) und (1$\bar{1}$0) den Kristall mit gut entwickelten Flächen; ihre entsprechenden parallelen Flächen sind mangelhaft ausgebildet. Die Flächenausbildung ist die am Muskovit gewöhnliche.

In die Fläche (001) ist ein 6 mm langes und 0,6 mm dickes Turmalinkriställchen zur Hälfte eingewachsen. Die Hauptachse des Turmalins läuft parallel zur Fläche (010), bzw. zur Symmetrieebene und senkrecht auf die optische Achsenebene. In die Fläche (001) sind ausserdem auch noch zwei kleine Quarzkristallbruchstücke eingewachsen.

Die physikalischen Eigenschaften sind die für den Muskovit kennzeichnenden.

ADULAR VON FELSŐBÁNYA (BAIA SPRIE).

Der Adular kommt in den Erzgängen der Karpathen recht selten vor, bzw. er wurde ziemlich selten erkannt. Das hängt in gewissem Masse mit der Anspruchslosigkeit des Minerals zusammen. *Mauritz* (10) schrieb zutreffend: »Valószínűnek tartom, hogy az adulár az andeziteknek még számos hazai érctelérében is megtalálható lesz: az igénytelen külsejű ásvány könnyen érthetőleg nem részesült kellő figyelemben« (p. 39.).*

* »Ich halte es für wahrscheinlich, dass der Adular noch in zahlreichen einheimischen Erzgängen der Andesite auffinden sein wird; dem Mineral wurde wegen seines anspruchslosen Äusseren leicht verständlich nicht die nötige Beachtung geschenkt.« (Übersetzung von *L. Tokody*).

Die ersten Angaben über den Adular von Felsőbánya stammen noch von *Krenner* (11). Er untersuchte die Wolframitkristalle und erwähnte, neben Schwefelkies, als Begleitmineral und jüngste Ausscheidung den Adular mit den Formen (110), (10$\bar{1}$) und selten noch (001). Der nähere Fundort dieser Kristalle ist Levesbánya.

S. Koch (12) beschäftigte sich mit der Kristallographie des Proustits und Pyrargyrits von Felsőbánya. Er stellte die folgende Paragenese fest: Adular, Quarz, Zinkblende, Schwefelkies und Dolomit. Er untersuchte unter den erwähnten Mineralien auch Adular und beobachtete an den wasserhellen, kurzprismatischen Kristallen die Formen (001), (010), (110), (10$\bar{1}$).

Im Jahre 1944 brachte ich eine Stufe aus den Erzgängen von Felsőbánya mit. An dieser stellte ich folgende Mineralien fest: Quarz, Pyrargyrit und Adular.

Die von mir untersuchten Adularkristalle kommen in einer anderen Paragenese vor, als die von *Krenner* und *Koch*. Bemerkenswert ist es aber, dass trotz des Paragenesenunterschieds die Formenentwicklung meiner Kristalle mit der von *Koch* übereinstimmt.

An sämtlichen von mir untersuchten Kristallen treten die folgenden Formen auf:

b(010)	x(10$\bar{1}$)
c(001)	m(110)

Die Hauptformen sind c(001), x(10$\bar{1}$) und m(110), dagegen ist b(010) untergeordnet. Der Typus der Kristalle ist briefkuvertähnlich.

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ÜBER EINEN APATIT

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Die basaltischen Gesteine des nógrád-gömörer Gebietes (Ungarn) gehören zu den Nephelinbasaniten und Nephelinbasanitoiden, (die ersteren mit Nephelin, die letzteren mit nephelinführendem Glas). Die Petrographie dieser Gesteine wurde in mehreren Mitteilungen behandelt. In diesen Arbeiten wird das Vorkommen von grossen Apatitkristallen nicht erwähnt. [1].

In der Umgebung von Somoskőújfalu (Ungarn), in etwa 5 km Entfernung gegen ONO von dieser Ortschaft, erhebt sich der Dobogó-Berg, ein Teil des basaltischen Gebietes von Medves. Das am Südabhang dieses Berges anstehende, etwas porös ausgebildete basanitische Gestein führt vereinzelt auch grössere Augitkristalle als porphyrische Einsprenglinge.

Die Augite sind meist 1—2 cm lang; seltener erreichen sie eine Länge von 3—4 cm. Sonst ist der Augit im Gestein — wie gewöhnlich — in kleinen Kriställchen gleichmässig veteilt.

Der grösste Teil der Apatitkristalle dieses Gesteines erscheint in winzig kleinen, farblosen Nädelchen, wie sie in allen Basalten gewöhnlich zu Hause sind. Selten findet man aber auch einen grossen Apatit, der — so zu sagen — als porhyrischer Einsprengling zum Vorschein tritt.

Dieser Apatit ist im Durchschnitt 10 mm hoch und 8 mm breit. Diese dicksäulenförmige Kristallgestalt besitzt die folgenden zwei Formen: Prisma erster Stellung $(10\bar{1}0)$ und Basis (0001). Der Kristall weist im Innern eine rissige Beschaffenheit und Querabsätze auf. Die letzteren verlaufen nahezu senkrecht zur Hauptachse des Kristalls. Die Farbe ist lichtgelb (kanariengelb), mit einem schwach grünlichen Stich. Im dickeren Dünnschliff lässt sich ein äusserst schwacher Pleochroismus beobachten: ω = blass gelb, ε = blass grünlich gelb.

Dieser grosse Apatit scheint ein seltener Gemengeteil zu sein. Grössere Apatitkristalle sind — bekanntlich — auch anderswo in basaltischen Gesteinen (Nephelinbasalt, Nephelindolerit) ab und zu beobachtet worden.

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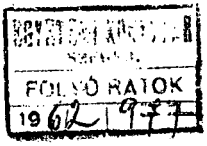

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